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# Heat Transfer Parameters of Cylindrical Catalyst Particles with Internal Voids in Fixed Bed Reactor Tubes

Dmitriy M. Rybak  
*Worcester Polytechnic Institute*

Michael David Ashman  
*Worcester Polytechnic Institute*

William T. Skene  
*Worcester Polytechnic Institute*

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# Heat Transfer Parameters of Cylindrical Catalyst Particles with Internal Voids in Fixed Bed Reactor Tubes

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A Major Qualifying Project Report

Submitted to the Faculty of the  
WORCESTER POLYTECHNIC INSTITUTE

in partial fulfillment of the requirements for the  
Degree of Bachelor of Science  
In Chemical Engineering

By

Michael Ashman

\_\_\_\_\_

Dmitriy Rybak

\_\_\_\_\_

William Skene

\_\_\_\_\_

Date: December 2009

Approved:

\_\_\_\_\_  
Dr. Anthony G. Dixon, Advisor

## **Abstract**

This study measured and analyzed fixed bed heat transfer parameters for different size and shape packing materials that might be used for steam methane reforming. These parameters were compared to those found in the literature for similar packing types with fair agreement. The monoliths and 4-hole cylinders gave better radial heat transfer than the raschig rings; however the monoliths are too brittle for industrial processes.

## Executive Summary

Steam methane reforming is a very important process for the industrial world and has many large scale applications. This process requires a very high operating temperature, high process flow rates, and also requires the presence of a catalyst. It is therefore necessary to optimize the process by finding a suitable catalyst shape and size that has a high surface area and a low pressure drop. The catalyst particle must be durable enough to withstand the process conditions, and it must also be able to transfer heat evenly and effectively to ensure uniformity throughout the process.

To more accurately study how different shaped particles transfer heat through fixed bed reactors, we employed a column with a heated air flow through the middle of the bed with a water cooled jacket to regulate temperature. The column was packed with different shaped packing materials and the heat dispersion was measured across the cross section of the column using an array of thermocouples that measured the temperature at several radial positions. The process was conducted using four different packing materials, each at four different bed depths and six different air flow rates. This process was conducted in a column with a diameter of 4 inches, and again in a column with a diameter of 2 inches.

Once the system was at steady state for each run, the temperature readings for each run were collected and analyzed. We used the Inlet Profile Plug Flow model to analyze the data and compared results for the different packing types. When fitted to the data, the IPPF model allowed us to determine which packing had the best heat transfer through the column. Overall, we found that the monoliths and 4-hole cylinders had better heat transfer characteristics than the raschig rings; however, we noted that the monoliths are too brittle to be considered for an industrial process as they are very prone to break and create dust (causing large problems in this type of setting).

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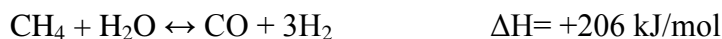
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## Introduction

In many cases, it is beneficial to convert natural gas into syngas ( $\text{H}_2$  and  $\text{CO}$ ) and then into a liquid through use of the Fischer-Tropsch process. The most common method used to convert natural gas into hydrogen is through conventional steam reforming (SR). Other methods similar to SR include Autothermal Reforming (AR) and Gas Heated Reforming (GHR). Commercial hydrogen production is also necessary for the production of ammonia, the production of methanol, as a reducing agent in metal ores, to increase the level of saturation in unsaturated fats and oils (such as margarine) and in hydrogen fuel cell research (New York Energy Research and Development Authority, 2009). The reaction taking place is shown below.



At high temperatures, above 700 degrees Celsius and with the use of a nickel catalyst, steam reacts with methane to produce hydrogen and carbon monoxide in fixed bed reactors. The process is highly endothermic, therefore this process consumes a lot of energy as heat. Thus, large amounts of energy are required to heat the reactors; over half of the plant's operating cost will be used for the syngas generation section alone. (US DOE, 2009) Production of hydrogen from methane has an efficiency of about 70%, and is the most common and cheapest method to produce hydrogen.

Fixed bed reactor tubes can have very high radial temperature gradients, and it is very important to ensure that there is effective heat transfer from the wall to the packing so that the reactor does not overheat and fail. A steam reformer facility usually has several hundred of these fixed bed reactor tubes and they each usually cost about \$6,000-\$7,000 due to the high process temperature that they are designed for. It is therefore very important to fully understand how the packing shape affects the radial heat transfer to maximize the lifespan of the reactors. Since altering the wall temperature of one of these tubes by 20 K can shorten the lifespan of the tubes from ten years to less than five, this can save the company millions of dollars over time (Dixon, 2009).

A lot of effort is therefore spent researching the best methods to heat the reactor tubes, so that the tubes are heated evenly and efficiently. There are several different approaches to the design of burners, including top, side and terraced wall firing for the steam reformers. Hundreds of burners are used to ensure that the heating is done in a uniform manner through the reformer.

In order to improve the interaction between the steam and the methane within the fixed bed, the reactor is filled with catalyst coated packing (Smirnov, 2003). The shape of the packing determines the amount of interaction between the steam and methane, and also determines the heat flow through the reactor. In order to control the process most efficiently, it is necessary to analyze how the shape and size of the packing alters the flow of heat through the reactor. In addition, the packing must function practically within the tube. The strength and durability of the packing are serious concerns and the packing must also be shaped properly to avoid sharp edges on the packing that lead to particle deterioration and dust that would contaminate the equipment.

Another vital concern in the development of a steam reformer is ensuring that the catalysts are loaded into the reactor tubes in a uniform manner (Stitt, 2005). If any tubes are packed tighter or less densely than another, then the reactant flow to the tubes which are in parallel would tend to have higher flows in some of the tubes than others which lead to hot spots and eventually system failure. To overcome this challenge, technicians perform highly accurate tests to ensure that the pressure drop across each tube for a given flow rate is as uniform as possible to ensure that the reactant flow does not tend towards certain reactor tubes over others.

There is a difficult tradeoff when deciding what kind of catalyst packing to use in a packed bed reactor. Catalysts are more effective with higher surface area, which would indicate that smaller particles with a larger surface area would be beneficial. But it is also true that larger packing particles require a lower pressure drop across the column, which allows for higher flow rates through the column. In addition, the tube to particle diameter ratio also determines the efficiency of the radial heat transfer (Stitt, 2005).

Research in the past regarding heat transfer modeling in packed beds is lengthy. Some of the first significant research was conducted by Coberly and Marshall in 1951, who were the first

engineers to use  $h_w$  and  $k_r$  as prior work had utilized  $k_r$  and  $k_r(r)$ . In 1972 DeWasch and Froment were the first to question the perceived dependence of  $k_r$  and  $h_w$  to bed height. In 1975, Gunn and Khalid were the first to introduce the concept of axial thermal dispersion, represented by the term  $k_a$ . In 1978, Bauer and Schünder developed what is today the standard model for  $k_r$ . Also in 1978, Dixon and Creswell introduced the theory that  $k_r$  and  $h_w$  were not height dependant. In 1985, Dixon showed that  $k_a$  had little physical basis, he also measured heat leaks through the calming section to eliminate height dependence. In 1988, Dixon reviewed  $Pe_r$  and  $Bi$  with a significant amount of heat transfer data. In 1993, Borkink, Borman and Westerterp introduced the Inlet Profile Plug Flow model to eliminate height dependence of the packing. In 1993, Martin and Niles found a relationship to describe  $Nu_w$ . In 1996, Landon, Hebert and Adams published results for different shaped packing materials. In 1997, Dixon looked at the effect that a low tube-to-particle ratio has on heat transfer for spheres.

There are many different shapes of packing used industrially for steam reforming, but there has been little research done exploring the heat transfer through fixed beds containing cylindrical packing with internal holes and domes. The main purpose of the team's research is to analyze the heat transfer across a fixed bed with different shaped packing. From these findings, optimal designs for catalyst particles can be suggested, which is a major step in the effort to develop more efficient steam reforming technology.

## Background

Heat transfer is categorized into three types: conduction, convection, and radiation. For these experiments, radiation does not apply as the temperatures were relatively low. Through research done over the years, models have been created using Fourier's law where conduction and convection parameters are strictly based on packing size and shape as well as fluid flow. The effective radial thermal conductivity,  $k_r$ , represents all mechanisms of heat transfer in the center of the bed (unaffected by wall effects). The wall heat transfer coefficient,  $h_w$ , is a parameter representing the increased heat transfer resistance close to the wall. Both of these parameters can be found from data using models such as the Plug Flow (PF) model or the Inlet Profile Plug Flow (IPPF) model.

In order to model the heat transfer using the equipment and model used, the following assumptions were made:

- The system was in steady state
- No reaction took place
- There is no radiation
- The pressure throughout the bed was constant
- No radial variation of the superficial gas velocity (Reynolds number)
- The wall temperature was constant and steady for each run
- The physical properties of the gas and solid were independent of temperature

## IPPF Model

The standard model used to analyze the temperature profile data acquired was the Inlet Profile Plug Flow (IPPF) model. The PF model uses the temperature of the inlet air as the inlet temperature; however, with the apparatus in the laboratory, it was impossible to determine the temperature of the air flowing into the packing at  $z = 0$  due to heat loss in the calming section. The IPPF model uses the first height temperature profile as the inlet temperature profile, and compares it to the temperature profiles at consequent heights.

The IPPF model is derived from the following dimensional energy balance:

$$G C_p \frac{\partial T}{\partial z} = k_r \left( \frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} \right)$$

With the following boundary conditions:

$$T = T_{z,o}(r) \text{ at } z = z_o$$

$$\frac{\partial T}{\partial r} = 0 \text{ at } r = 0$$

$$-k_r \frac{\partial T}{\partial r} = h_w (T - T_w) \text{ at } r = R$$

These equations are usually made dimensionless by defining the following:

$$\vartheta = \frac{(T - T_w)}{(T_o - T_w)}$$

$$y = \frac{r}{R}$$

$$x = z/R$$

$$Pe_R = \frac{G C_p R}{k_r}$$

$$Bi = \frac{h_w R}{k_r}$$

In this case, using the first measurement height at  $z=z_o$  as the inlet temperature, we get:

$$\zeta = \frac{z/L - z_o/L}{1 - z_o/L}$$

$$\frac{\partial \zeta}{\partial z} = \frac{1}{L} \frac{1}{1 - z_o/L}$$

So:

$$\frac{\partial T}{\partial z} = \frac{\partial T}{\partial \zeta} \frac{\partial \zeta}{\partial z} = \frac{1}{L} \frac{1}{1 - z_o/L} \frac{\partial T}{\partial \zeta}$$

Then:

$$G C_p \frac{1}{L} \frac{1}{1 - z_o/L} \frac{\partial \vartheta}{\partial \zeta} = k_r \frac{1}{R^2} \left( \frac{\partial^2 \vartheta}{\partial y^2} + \frac{1}{y} \frac{\partial \vartheta}{\partial y} \right)$$

i.e.

$$\frac{\partial \vartheta}{\partial \zeta} = \frac{k_r L}{G C_p R^2} (1 - z_o/L) \left( \frac{\partial^2 \vartheta}{\partial y^2} + \frac{1}{y} \frac{\partial \vartheta}{\partial y} \right)$$

Define:

$$Bo_{h,r}^\vartheta = \frac{G C_p R^2}{k_r L}$$

$$\theta = \frac{\vartheta}{\vartheta_o}$$

Where:

$$\vartheta_o = \frac{(T|_{r=o} - T_w)}{(T_o - T_w)}$$

Gives:

$$\frac{\partial \theta}{\partial \zeta} = \frac{1}{Bo_{h,r}^\vartheta} \left( 1 - \frac{z_o}{L} \right) \left( \frac{\partial^2 \theta}{\partial y^2} + \frac{1}{y} \frac{\partial \theta}{\partial y} \right)$$

With boundary condition:

$$-k_r \frac{\partial T}{\partial r} = h_w (T - T_w)$$

This can be made dimensionless into:

$$-k_r \frac{1}{R} \frac{\partial \vartheta}{\partial y} = h_w \vartheta$$



This may be reduced to the following:

$$\frac{\partial \vartheta}{\partial y} + Bi\vartheta = 0$$

And then:

$$\frac{\partial \theta}{\partial y} + Bi\theta = 0$$

Assume a parabolic profile at  $z = z_o$ :

$$T = T|_{r=0} + A'r^2$$

Where  $A' < 0$  for cooling.

$$\frac{(T - T_w)}{(T_o - T_w)} = \frac{(T|_{r=0} - T_w)}{(T_o - T_w)} + \frac{A'}{T_o - T_w} R^2 y^2$$

$$\vartheta = \vartheta_o + \frac{A'R^2}{T_o - T_w} y^2$$

So:

$$\theta = \frac{\vartheta}{\vartheta_o} = 1 + \frac{A'R^2}{(T_o - T_w)\vartheta_o} y^2 = 1 - Ay^2$$

Where  $A = \frac{A'R^2}{(T_o - T_w)\vartheta_o} > 0$  always.

The IPPF model is incorrect as given by Borkink (1993). It may be made correct by defining:

$$Bo_{h,r}^{\theta}(\text{corrected}) = \frac{GC_p R^2}{k_r L} \frac{1}{1 - z_o/L} = Bo_{h,r}^{\theta} \frac{1}{1 - z_o/L}$$

Convert into present variables using corrected  $Bo_{h,r}^{\theta}$ :

$$Bo_{h,r}^{\theta} = Pe_R \frac{R^2}{Ld_p} \frac{1}{1 - z_o/L} = Pe_R \frac{R}{L} \frac{1}{1 - x_o R/L}$$

$$\zeta = \frac{x^{R/L} - x_o^{R/L}}{1 - x_o^{R/L}}$$

$$\frac{\partial \zeta}{\partial x} = \frac{R}{L} \frac{1}{1 - x_o^{R/L}}$$

$$\begin{aligned} \vartheta &= \frac{(T - T_w)}{(T_o - T_w)} / \frac{(T|_{r=o} - T_w)}{(T_o - T_w)} \\ &= \left[ \frac{(T_w - T_o)}{(T_w - T_o)} - \frac{(T - T_o)}{(T_w - T_o)} \right] / \left[ \frac{(T_w - T_o)}{(T_w - T_o)} - \frac{(T|_{y=o} - T_o)}{(T_w - T_o)} \right] \\ &= (1 - \vartheta)/(1 - \vartheta_o) \end{aligned}$$

Equations become:

$$\frac{\partial \vartheta}{\partial x} = \frac{1}{Pe_R} \left( \frac{\partial^2 \vartheta}{\partial y^2} + \frac{1}{y} \frac{\partial \vartheta}{\partial y} \right)$$

With:

$$\vartheta = 1 - (1 - \vartheta_o)(1 - Ay^2) \text{ at } x = x_o$$

$$\vartheta \text{ finite at } y = 0.$$

$$\frac{\partial \vartheta}{\partial y} + Bi(\vartheta - 1) = 0 \text{ at } y = 1$$

This reduces to the conventional PF model if  $x_o = 0$ ,  $A = 0$ , and  $\vartheta_o$ . The solution is:

$$\begin{aligned} \vartheta &= 1 - 2(1 - \vartheta_o) \sum_{i=1}^{\infty} \left\{ \frac{[(Bi\lambda_i^2 + 4ABi - 2A\lambda_i^2 - ABi\lambda_i^2)J_o(\lambda_i y)]}{\lambda_i^2 [\lambda_i J_o(\lambda_i) + BiJ_i(\lambda_i)]} e^{\left[ -\frac{\lambda_i^2}{Pe_R}(x - x_o) \right]} \right\} \\ &= 1 - 2(1 - \vartheta_o) \sum_{i=1}^{\infty} \left\{ \frac{(Bi + 4ABi/\lambda_i^2 - 2A - ABi)J_o(\lambda_i y)}{[Bi^2 + \lambda_i^2]} \frac{J_o(\lambda_i y)}{J_o(\lambda_i)} e^{\left[ -\frac{\lambda_i^2}{Pe_R}(x - x_o) \right]} \right\} \end{aligned}$$

## Dimensionless Parameters

The Reynolds number (Re) is a dimensionless fluid mechanics parameter that is directly proportional to the superficial velocity of the fluid. The equation for the Reynolds number of the air flowing through a fixed bed is:

$$Re = \frac{v_s \rho d_p}{\mu}$$

Where  $v_s$  is the superficial velocity of the air in ft/min,  $\rho$  is the density of the air,  $d_p$  is the diameter of the particle in inches, and  $\mu$  is the dynamic viscosity of the air

The Peclet number ( $Pe_r$ ) is given by the following equation:

$$Pe_r = \frac{Re * Pr}{k_r / k_f}$$

Assuming  $k_r/k_f$  is linearly related to Re, and Pr is assumed to be constant at 0.71 for air at 60 °C:

$$Pe_r = \frac{Re * Pr}{m * Re + b}$$

Where m and b are the slope and intercept respectively of the line fitting our  $k_r/k_f$  versus Re data. The Peclet number should increase rapidly for a low range of Reynolds numbers and stay relatively constant with respect to Reynolds number after reaching a certain velocity. This is because when Re is low, the intercept (b) has more of an effect on  $Pe_r$ .

The Biot Number (Bi) is a relationship between the wall heat transfer coefficient and the thermal conductivity of the packing. It is given by the following equation:

$$Bi = \frac{h_w R}{k_r} = Nu_w \frac{R/d_p}{k_r / k_f}$$

Since  $k_r$  is assumed to be linearly proportional to the Reynolds number, and  $h_w$  increases at a slower rate with Reynolds number, we would assume the Biot number to decrease with respect to the Reynolds number. If  $Nu_w$  was linearly proportional to  $Re$ , the Biot number would decrease linearly, but theoretically  $Nu_w$  should fit the equation:

$$Nu_w = Nu_w^o + ARe^B Pr^C$$

Where A, B, and C are experimentally determined constants. With only six data points, it is impossible to create an accurate fit line with this model for  $Nu_w$ , so we just assume that the Biot number will decrease with respect to Reynolds number.

The dimensionless parameter N is a ratio of tube diameter to particle diameter.

$$N = \frac{2 * R}{d_p}$$

Several studies completed have given various parameters that must be met in order to receive accurate data when using models such as the IPPF. Many of these studies suggest that the first bed depth used should be at least 1.5 times the diameter of the column (Borkink & Westerterp, 1993). This being said, the optimum first height for the 4 inch column would be 6 inches. With the limited number of particles available, the first height chosen was 4 inches, which could have had an effect on the accuracy of our data.

## Methodological Procedure

### Equipment

The apparatus used in a cooling experiment with a fixed bed reactor tube is shown in Figure 1.

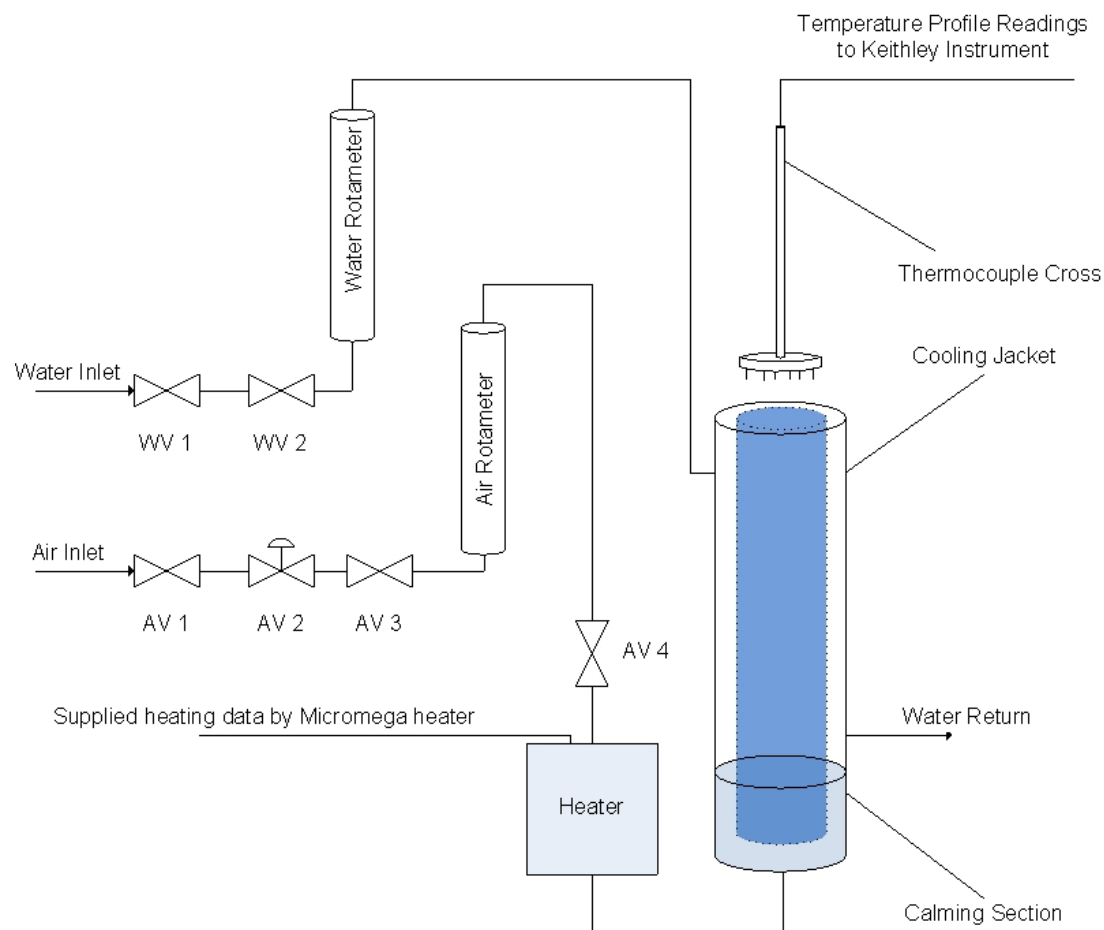


Figure 1: Schematic of Laboratory Apparatus

### Columns

Each column used was constructed using two concentric brass tubes to form an annular space between the two (water jacket). The columns were attached to calming sections using four screws. Screws used in the 4 inch column were constructed from nylon, whereas the screws used in the 2 inch column were constructed from steel. The calming sections were constructed out of nylon and filled with  $\frac{1}{4}$  inch steel spheres. The steel spheres were separated from the test packing by a circular piece of steel mesh. The calming section allowed for the airflow to be evenly

dispersed over the cross-section of the column. The columns had internal diameters measuring 2 inches (50.8mm) and 4 inches (101.6mm).

### Determining Reynolds number of inlet air flow

To measure the flow rate of the air entering the column, it was passed through a ½-27-G-10 rotameter tube with a ½-GSVT-48A float. Depending on the pressure of the air, different amounts of air would flow through the rotameter at 100% of the total flow. These values are listed in Table 1.

**Table 1: Flow data for a ½-27-G-10 rotameter tube with a ½-GSVT-48A float**

Gauge pressure	MAX	Gauge Pressure	MAX
0	44.9	26	2.94
1	4.73	27	2.91
2	4.6	28	2.88
3	4.46	29	2.84
4	4.34	30	2.8
5	4.23	31	2.78
6	4.13	32	2.75
7	4.03	33	2.72
8	3.94	34	2.69
9	3.86	35	2.67
10	3.78	36	2.64
11	3.7	37	2.61
12	3.64	38	2.59
13	3.57	39	2.56
14	3.51	40	2.54
15	3.45	41	2.52
16	3.39	42	2.49
17	3.34	43	2.47
18	3.28	44	2.45
19	3.23	45	2.43
20	3.19	46	2.41
21	3.15	47	2.39
22	3.1	48	2.37
23	3.06	49	2.35
24	3.02	50	2.34
25	2.98	51	2.32

The volumetric flowrate of air flowing through the column (SCFM) was found using the following equation:

$$SCFM = \% * Max * \left( \frac{14.7 + psig}{14.7} \right)$$

Where % is the percentage read on the rotameter tube for air, Max is the maximum flow rate (at 100% on the rotameter, which can be found in Table 1) for the rotameter tube and float used, and psig is the pressure read off of the pressure gauge downstream from the rotameter. The percentage of airflow is read from the “knife-blade edge” of the float (which is at the widest diameter of solid on the float).

Since the column area, and the density and viscosity of air were assumed constant for air flowing through the column, the Reynolds number was then determined using the following equation derived for each column:

$$Re = C * SCFM * d_p$$

Where C is 380 for the 2 inch column and 95 for the 4 inch column.

## Packings

Four different packing shapes were examined throughout the course of the project:

- ½ inch ceramic spheres
- Raschig rings
- Johnson Matthey K-57 4-hole pellets (4-hole cylinders)
- Monoliths

The ½ inch ceramic spheres measured 12.7mm in diameter and were porous in nature. There have been many studies completed using spheres with N=8 (in our case, the spheres were used in the 4 inch column only). By comparing our data to these studies, we could verify that our data is consistent with past experiments.

Raschig rings in this study are brittle non-porous ceramic cylinders with a single hole in the middle. The rings we used had heights and outer diameters equal to 13mm, with a wall thickness of 3mm. There have also been some studies with raschig rings with various values of N, which

allows us to do a comparison to verify our data is relatively consistent and will give values for the N we used.

The Johnson Matthey K-57 4-hole pellets were porous ceramic 4-hole cylinders. These were 17mm tall, with an outer diameter of 14mm and hole diameters of 4mm. Packings such as these are typical support for steam reforming.

The monoliths are ceramic cylinders, measuring 7mm tall with a diameter of 6mm and a hole density of 0.563holes/mm<sup>2</sup> (56.3 holes/cm<sup>2</sup>). The monoliths used in this study are illustrated in Figure 2.

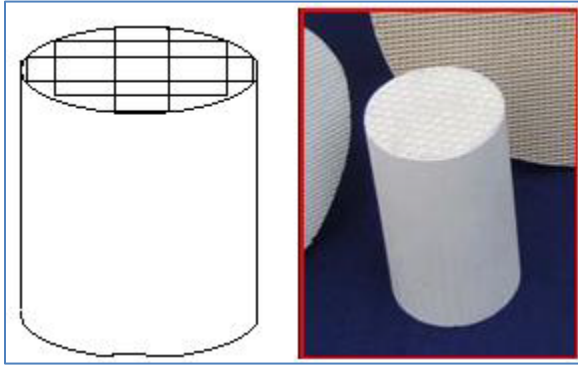


Figure 2: Monoliths (<http://www.made-in-china.com/showroom/hisina88/product-detailKoXmSOuGZzVf/China-Automobile-Exhaust-Catalyst-Carrier.html>)

The Inlet Profile Plug Flow model requires a particle diameter to complete calculations. This diameter is referred to as the equivalent spherical diameter. In other words, this is the diameter required in a sphere to obtain the same volume as the particle (without accounting for any internal voids). The three non-spherical packings were all cylindrical in shape, so the equivalent spherical diameter could be found using the following equation:

$$\pi \frac{d_c^2}{4} h = \frac{4}{3} \pi \frac{d_p^3}{8}$$

Where  $d_c$  is the diameter of the cylinder and  $d_p$  is the equivalent spherical diameter.

Table 2 lists the equivalent spherical diameters of the packings, as well as the ratio of each column to said diameters.



**Table 2: Packing to column diameter ratios**

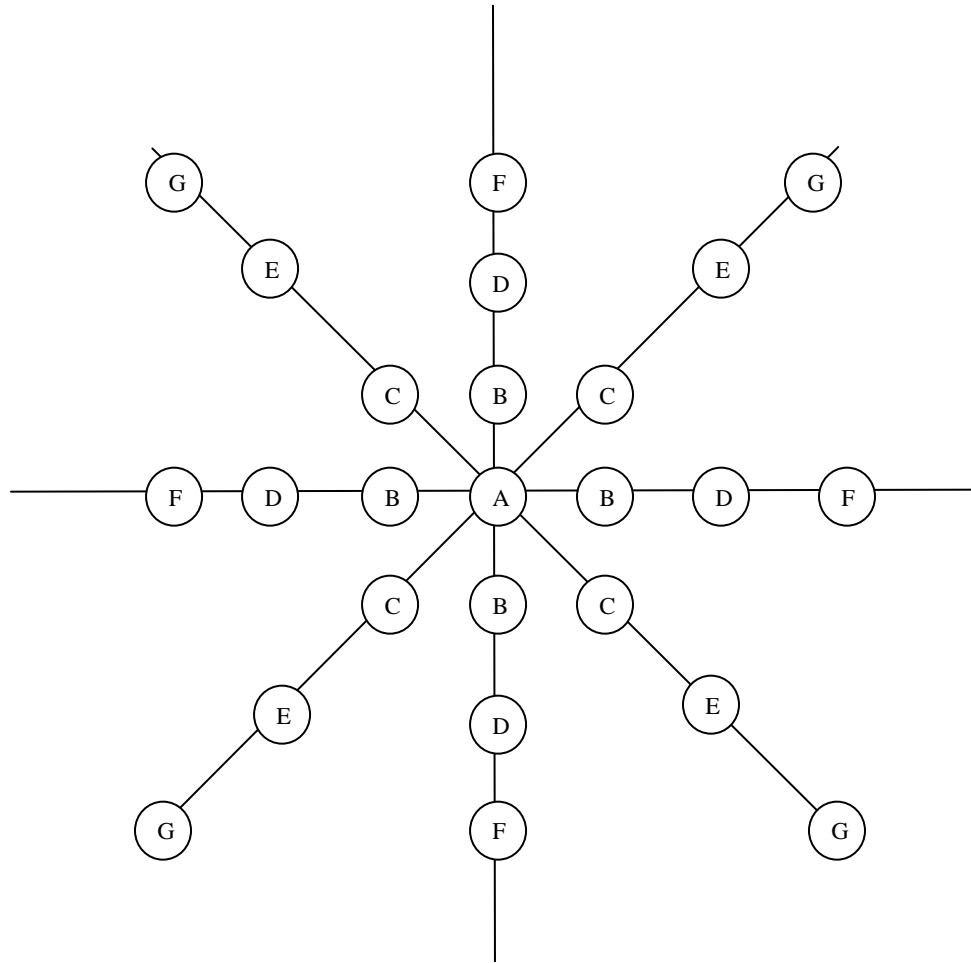
Packing	Equivalent Spherical Diameter	N in the 2 inch column	N in the 4 inch column
Spheres	12.7	--	8.0
Raschig Rings	14.5	3.5	7.0
JM 4-hole cylinders	17.4	2.9	5.8
Monoliths	7.3	7.0	14.0

### **Micromega Heater**

The heater used to heat the inlet air flow was controlled by an Omega Micromega CN77000 Series Controller. Complete instructions to use the controller may be found on Omega's website at <http://www.omega.com/Manuals/manualpdf/M2491.pdf>. The purpose of the heater in this experiment is to heat the inlet air to approximately 100 °C. There are two settings that may be altered to acquire this: the desired temperature and the rate at which it takes to get to the desired temperature (ramp and soak). Our desired temperature was set to 100 °C. The rate will affect how long it will take the system to reach equilibrium. Since it is impossible to get the temperature of the air to be exactly 100 °C at all times, the heater heats until the air is 100 °C and hinders heating until the flow is cooled below 100 °C, in which case it will start heating again. This creates an oscillation in temperature over time, and after infinite time the temperature will approach 100 °C. With a faster heating rate, the air flow will reach 100 °C sooner; however, larger oscillations will occur. Conversely, with a slower heating rate, the air flow will reach 100 °C slower, but smaller oscillations will occur. The ramp and soak configuration was disabled, which allows the heater to reach the desired setting as fast as possible.

### **Thermocouples**

A diagram of the thermocouple cross can be seen in Figure 3.



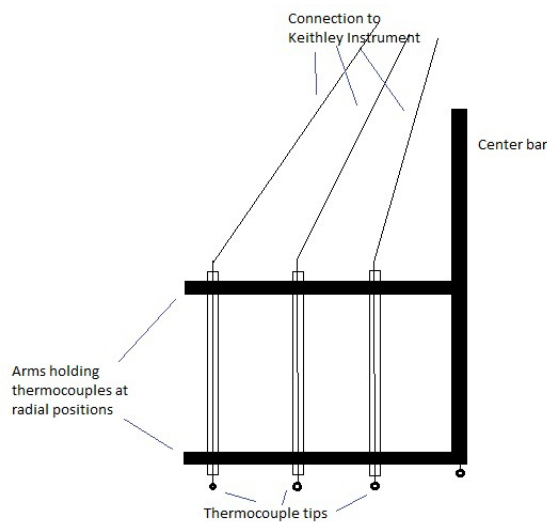
**Figure 3: Thermocouple Cross**

The thermocouple cross consisted of two sets of four arms set 45 degrees apart from each other. Each arm had three thermocouples attached, measured at different radii. The radii for each thermocouple in millimeters are shown in Table 3.

**Table 3: Radial position of each thermocouple represented in Figure 2 in both columns**

Thermocouple	2 inch column (mm)	2 inch column (dimensionless)	4 inch column (mm)	4 inch column (dimensionless)
A	0	0.00	0	0.00
B	8.5	0.33	9.5	0.19
C	12	0.47	19	0.37
D	15	0.59	28	0.55
E	18	0.71	41	0.81
F	21.5	0.85	44	0.87
G	24	0.94	48	0.94

Not counting the center thermocouple, data is collected by 24 thermocouples at six different radial positions. By rotating the thermocouple cross 45° and taking a second set of readings, 48 thermocouple readings are recorded (i.e. eight angular replicates of six radial positions). This allows for a more accurate measurement of the average temperature of each radial position across the cross-section of the column. The thermocouples were held in place by thin plastic tubing (like a small straw) connected to two nylon bars (each arm consisted of two bars) with holes drilled at the same radial position. A side view of the thermocouple cross (one of the eight arms) is shown in Figure 4.



**Figure 4: Side view of one arm in the thermocouple cross**

Consecutive thermocouples used in these experiments are placed at radii very similar to the ones before and after (within 3mm). These thermocouples must be verified before inserting the thermocouple cross into the column each time to ensure that they are not bent, as reading temperatures from 1 or 2 mm different than expected will yield faulty results.

Three thermocouples were placed in the cooling water jacket. These were placed 7.5, 24.5, and 37.5 mm from the bottom of the column. All three were averaged for each set of data in order to determine the wall temperature.

### Temperature Profile Collection

The data acquisition device used was a Keithley Integra Series 2700 Datalogger. This device combines functions of DMM, switch system, and datalogger, and allows for the collection of

thermocouple data for up to 200 channels (35 of these channels were utilized in this experiment). The device worked in conjunction with ExceLINX (an add-in designed for Microsoft Excel) to collect data over time.

To install ExceLINX, the program file ExceLINX.xla must be placed in the Excel Add-ins folder. This can be found in explorer: C→Documents and Settings→All users→Application Data→Microsoft→Add Ins. Then in Excel, go to Tools→Add In. Click browse and select ExceLINX.xla from the menu. Now the ExceLINX tab should be available from the menu at the top of Excel.

The Keithley instrument must now be configured to use with the ExceLINX program. Start by turning on the Keithley instrument. Then, at the top of the Excel screen, select ExceLinx→DMM Config and complete as shown in Figure 5.

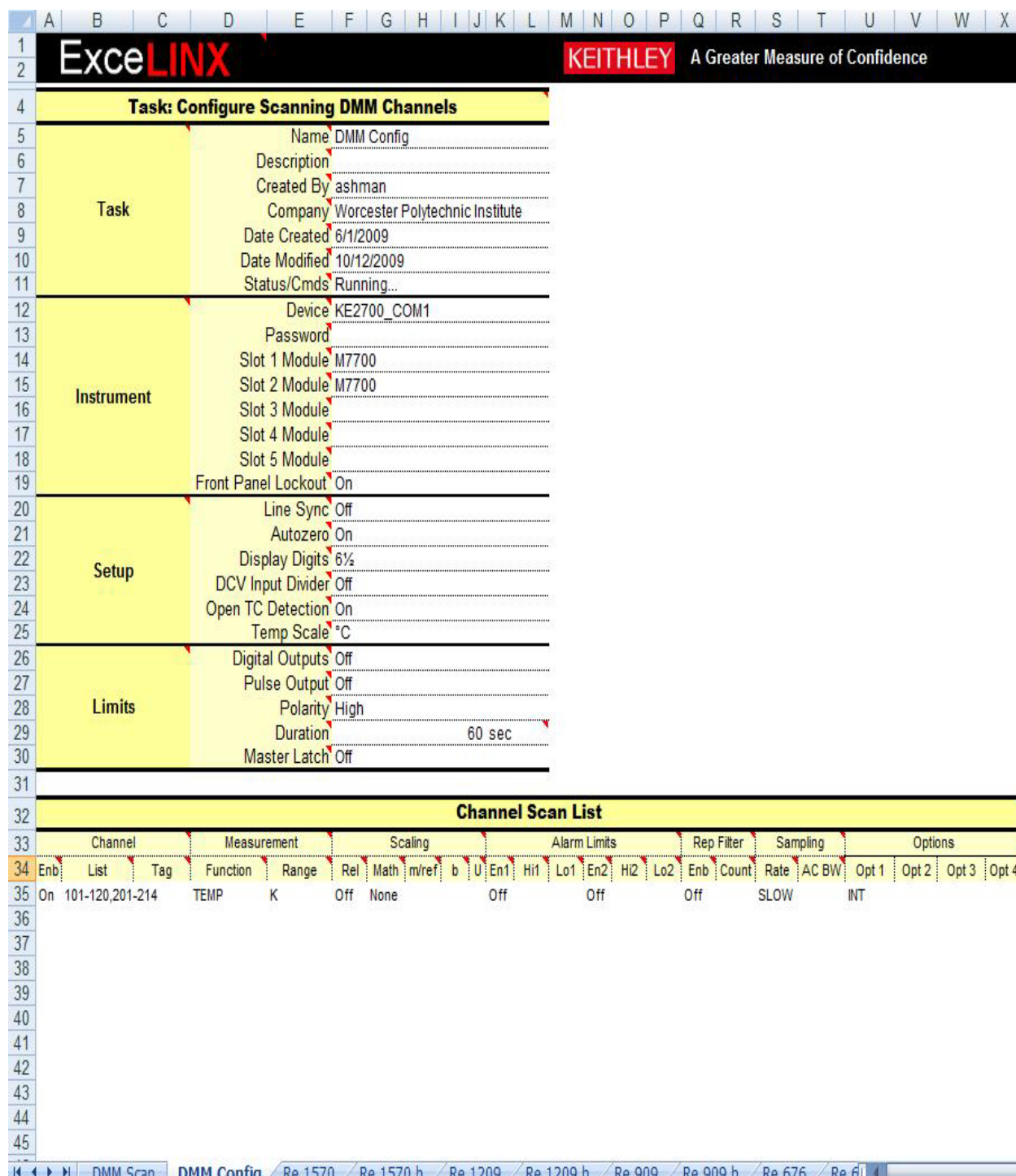


Figure 5: DMM Config Screenshot

Then click ExceLINX→DMM Scan and complete as shown in Figure 6.

	A	B	C	D	E	F	G	H	I	J
1	<b>ExceLINX</b>									
2	<b>KEITHLEY</b> A Greater Measure of Confidence									
4	<b>Task: Scan DMM Channels</b>									
5	<b>Task</b>	Name	DMM Scan							
6		Description								
7		Created By	ashman							
8		Company	Worcester Polytechnic Institute							
9		Date Created	6/1/2009							
10		Date Modified	10/12/2009							
11	Status/Cmds	Task stopped successfully								
12	<b>Configuration</b>	Worksheet	DMM Config							
13	<b>Trigger</b>	Model	Scan							
14		Source	Immediate							
15		Delay	Auto	sec						
16		Reading Count	INF							
17		Timer	0.1	sec						
18		Monitor	None							
19	Monitor Limits	None								
20	<b>Data Location</b>	Worksheet	DMM Scan							
21		Starting Col	A							
22		Starting Row	1							
23		Organize By	Rows							
24		Autoincrement	Use one table							
25		Auto Wrap	On							
26	Log File									
27	Format	Delimited text (comma)								
28	<b>Data Display</b>	Add Channel Tags	Yes							
29		Add Channels	Yes							
30		Add Units	No							
31		Scroll Display	No							
32		Limits	None							
33		Timestamp	None							
34	Update Interval	100	msec							
35										
36	<b>Task Data</b>									
37										
38										
39										
40										
41										
42										
43										
44										
45										

Figure 6: DMM Scan Screenshot

To record data, select the dropdown menu “Status/Cmds”, select “Start”, and then press enter. Data will be recorded in the selected sheet under “Worksheet”. To stop collecting, choose “Stop” from the “Status/Cmds” drop down menu and press enter very shortly after (almost instantaneously). Each column of data represents temperatures read at specific thermocouples

depending on their placement. In order to determine which thermocouple corresponded to each column, each thermocouple was tested by touching one by one with a finger-tip and checking ExceLINX to determine which column's temperature changed. The ambient temperature in the lab was usually close to 20 °C and after being touched by a finger-tip, the temperature reading for that thermocouple would read about 30 °C. There were five thermocouples removed from the column, but still connected to the Keithley instrument. Temperatures for these thermocouple connections read 9E31 on ExceLINX. After the channels on ExceLINX for these five thermocouples and the 25 thermocouples from the thermocouple cross were determined, the channels for the wall thermocouples and the inlet airflow needed to be determined. These were found by removing the connecting wires one by one and seeing which channel read 9E31.

## Procedure

In order to ensure a good fit for our data, six different Reynolds numbers were measured during each day of column operation, and four bed depths were taken for the packings in each column. A table of all runs completed during the course of the project is shown in Table 4.

**Table 4: Table of runs**

Column Diameter (mm)	Packing	Re Values	Bed Depths (mm)
101.6	½" spheres	75, 145, 195, 260, 335, 425	92, 150, 200, 225
101.6	Raschig Rings	85, 142, 190, 258, 341, 432	100, 150, 200, 250
101.6	JM 4-hole cylinders	102, 198, 268, 358, 460, 584	100, 150, 200, 250
101.6	Monoliths	43, 71, 95, 129, 171, 216	100, 160, 200, 260
50.8	Raschig Rings	350, 490, 676, 909, 1209, 1570	80, 150, 210, 255
50.8	JM 4-hole cylinders	409, 775, 1052, 1412, 1822, 2275	80, 150, 200, 265
50.8	Monoliths	170, 323, 439, 589, 760, 948	95, 155, 210, 260

It has been determined that the minimum packing depth should be 1.5 times the column diameter to let the parabolic temperature profile develop (Borkink & Westerterp, 1993). This must be

done so that the IPPF model would fit the data (requires parabolic temperature profiles). Although the minimum packing depth was determined to be 1.5 times the column diameter, this could not be done with some of the packings in the 4 inch column due to a lack of packing quantity. To ensure enough deviation in bed depths, and keeping consistency throughout the runs, runs in the 4 inch column started at a height close to 4 inches (only 1 times the column diameter).

### Operating the Column

1. If running the tower for the first time, carefully unscrew and remove the top section of the tower.
2. Fill the calming section of the tower with metal spheres and cover the calming section with a wire mesh to keep it separate from the test packing.
3. Re-attach the top section of the tower.
4. Measure the inside height of the empty tower and record the value.
5. Add the desired amount of the packing you are working with to the tower. To do this properly you should:
  - a. Add the packing little by little and gently compact it from the top after each addition to assure for uniformity of packing.
  - b. Regularly measure the height of the empty space left in the column (space from packing to top) to ensure that you do not surpass your desired packing height.  
(Packing height=height of empty tower - empty space left)
6. Set up the thermocouple cross so that once it is inside the tower the thermocouples are just above the packing (no more than 3-6 mm). To do this measure the distance from the cross bar that will support the cross to the bottom of the thermocouples and set it so that this distance is slightly less than the empty space remaining in the tower. Before lowering the thermocouple cross into the column each time, straighten all thermocouple tips and ensure that they are centered with the guide tubes.



7. Carefully lower the thermocouples cross into the tower. If you feel it hit the top of the packing remove it from the tower, re-adjust the height of the cross bar, and straighten all thermocouple tips.
8. Open ExceLINX. If you have not yet set up ExceLINX as described in the ExceLINX section take the time to do so now.
9. Turn on and configure the Keithley 2700 Multimeter/Data Acquisition System.
10. Prior to starting any flows make sure that all the valves are in the fully shut position. For Water Valves (WV) 1&2 as well as Air Valves (AV) 1&3 this means that the valve is perpendicular to the tube on which it is located. For AV 2 this means that the valve is turned fully in the CLOCKWISE direction.
11. Slowly fully open WV 1.
12. Slowly open WV 2 until the rotameter reads a flow of 80%.
13. Slowly fully open AV 1. At this point if the pressure gauge following AV 1 is showing a non-zero pressure reading it means that AV 2 is not fully shut, in which case take the time to fully shut it now.
14. Slowly fully open AV 3.
15. If you are running the column for the first time adjust AV 4 so that at the desired setting it allows for a full range of desired Re values to be obtained using only AV 2 to adjust the flow. The following things should be noted:
  - a. Once this has been set AV 4 should not be touched in further runs to ensure for uniformity of values.
  - b. Never allow the rotameter to reach or exceed 100%.
  - c. The pressure gauge that should be used to calculate the Re values is the one that is right before entering the heater (i.e. downstream from the rotameter).

16. Turn on the heater. If you have not yet set up the Omega Micromega heating control unit take the time to do so now.
17. Set the flow to the desired amount by adjusting only AV 2.
18. Using ExceLINX start recording the data.
19. Once the system has reached steady state stop the data recording. This should take anywhere from 1 to 3+ hours depending on the flow rate, packing height, and if it's the first run of the day.
20. Carefully rotate the thermocouple cross 45 degrees. The top of the column has markings for 0 and 45° in which the cross is to be lined up with to accurately measure the 45° rotation.
21. Commence recording data in ExceLINX. Make sure to do this in a different Excel sheet than the one used for the previous data.
22. Once the system has once more reached steady state stop the data recording. This should only take about 5min after the angular rotation.
23. Repeat Steps 17-22 for all desired Re values.
24. Shut the system down in the reverse order that you started it in.

## Safety Precautions

The spheres, raschig rings, and monoliths were all inert ceramic packings. However, the 4-hole cylinders contained a white powdered metal on the surface of each particle. Labels on the container provided by Johnson and Matthey warn that exposure may cause irritation. Although it has not been classified as hazardous, it is good practice to minimize exposure to this metal. In order to do so, gloves were worn at all times when handling the 4-hole cylinders. Also, in order to prevent inhalation, the first set of runs performed with this packing was done using the largest bed depth. Once packed for the first time, painter's masks and safety glasses were worn and then the air was set to the highest flowrate achievable. Visibly, no dust came from the column at the highest flowrate, so the air was left at a high flowrate overnight so that any particles that did get

blown up were not breathed in by anyone. No dust was visible anywhere in the lab the next day; a miniscule amount was actually loose enough to be blown off the particles.

The heater would reach temperatures of 115° C, and even though was insulated on all sides, could cause minor burns if touched. These surfaces were not touched for any reason, and if a particle or other object were to fall on top of the heater, it was immediately removed with pliers.

### Data file format for model fitting

The IPPF model was fit to the temperature profile readings using an existing program written in FORTRAN. In order to run the program, it was required to create a text file in the correct format with data collected. A template of this format is shown in Table 5. In order to smooth the steady state thermocouple temperatures, the last five time values in our ExceLINX data collection sheet were averaged.

**Table 5: FORTRAN input template**

# of Profiles	# of Radial Positions	# of wall Temperature Readings	# of Angles		
Column Diameter	Particle Diameter				
Radius 1	Radius 2	Radius 3	Radius 4	Radius 5	Radius 6
Reynolds Number	Bed Depth	Angle of Rotation			
Inlet Temperature					
Radius 1 Thermocouple 1	Radius 1 Thermocouple 2	Radius 1 Thermocouple 3	Radius 1 Thermocouple 4		
Radius 2 Thermocouple 1	Radius 2 Thermocouple 2	Radius 2 Thermocouple 3	Radius 2 Thermocouple 4		
Radius 3 Thermocouple 1	Radius 3 Thermocouple 2	Radius 3 Thermocouple 3	Radius 3 Thermocouple 4		
Radius 4 Thermocouple 1	Radius 4 Thermocouple 2	Radius 4 Thermocouple 3	Radius 4 Thermocouple 4		
Radius 5 Thermocouple 1	Radius 5 Thermocouple 2	Radius 5 Thermocouple 3	Radius 5 Thermocouple 4		
Radius 6 Thermocouple 1	Radius 6 Thermocouple 2	Radius 6 Thermocouple 3	Radius 6 Thermocouple 4		
Wall Thermocouple 1	Wall Thermocouple 2	Wall Thermocouple 3			

One text file could be made for all packing depths and Reynolds numbers for each packing in each column. Therefore, when using this template, the top three lines were only needed once at the top of the file. To indicate the end of the file, a “-1” was inserted on the last line to notify the program to stop. A sample text file is shown in Appendix A: Sample Fortran Input Text File (for JM 4-hole cylinders in the 2 inch column).

## Fitting the IPPF model to collected data

In order to obtain a solution for the program in FORTRAN, the IPPF program uses iterations to minimize the sum of squares function given below:

$$\min(Pe_r, Bi) S = \sum (T_{calc} - T_{measured})^2$$

Where the measured temperatures are taken at each angular and radial position for the different bed depths. The temperatures are made dimensionless by:

$$\vartheta = \frac{(T - T_w)}{(T_o - T_w)}$$

The IPPF model can now be solved analytically. The program performs two sets of functions: first solving the partial differential equation, and the second minimizing the sum of squares shown above as a function of input parameters. The sequence in which this is performed is:

1. Guess  $Pe_r$  and  $Bi$
2. Solve partial differential equation
3. Form  $S$
4. Is  $S$  at a minimum? If not, start back at 1 and reiterate

For the IPPF model,  $Pe_r$  and  $Bi$  vary for each packing and initial values are needed before running the program. . The Biot number was assumed to be anywhere between 1 and 2, so was usually guessed to be 2 (unless iterations were not able to be completed). The Peclet number has been suggested from past research to be about 10 for spheres, and 6 for rings.

The function for the sum of squares is then solved using the guessed values for  $Pe_r$  and  $Bi$ . This sum is ended dependent on the user's desired accuracy.

The sum of squares function is in the category of target functions  $S(\underline{x})$  with  $\underline{x}=(x_1, x_2, \dots, x_n)$

$$S(\underline{x}) = \sum_{i=1}^m f_i(\underline{x})^2 = \underline{f}^T \underline{f}$$

Where  $m$  is the number of data points and  $n$  is the number of parameters to be estimated. Then, a  $m \times n$  Jacobian matrix is formed with the first partial derivatives:

$$J_{ij}(\underline{x}) = \left( \frac{\partial f_i}{\partial x_j} \right)_{\underline{x}}$$

Using this, the gradient of S is given by:

$$G = 2J^T J + 2 \sum_{i=1}^m f_i K^i$$

Where  $K^i$  is the matrix of second derivatives of  $f_i$  (under some conditions, may be zero). S is minimized by the Levenberg-Margardt algorithm. The accuracy of this solution is then determined by the accuracy of the radial temperature measurements used in the iterations. The approximate error ( $\underline{E}$ ) is then obtained by solving the following:

$$\left( J(\underline{x})^T J(\underline{x}) \right) \underline{E} = -J(\underline{x})^T \underline{f}(\underline{x})$$

The second derivative of the target function must be available to approximate  $\underline{x}$ . The actual method for calculating  $\underline{E}$  depends on which of  $J$ ,  $J^T J$ , or  $(J^T J)^{-1}$  is available. The accuracy of  $x_i$  values is determined by observing how  $x_i$  changes over iterations. Values for  $x_i$  should not change over the last few iterations (Van Dongeren, 1994).

Although these iterations allow the determination of accurate values for Peclet and Biot numbers, it is important to determine confidence intervals for these parameters. As long as the hessian of S is available in some form, variances may be calculated. If J is known,  $G=2J^T J$  can be evaluated. Letting H be the inverse of G and S be the sum of squares, then an estimate of the uncertainty in the  $i^{\text{th}}$  parameter  $x_i$  is:

$$var(x_i) = \frac{S}{m-n} H_{ii}$$

Now that the variance is known, the values for a t-distribution with m-n degrees of freedom can be used to give 100(1- $\beta$ ) confidence intervals for  $\underline{x}$ :

$$x_i - \sqrt{var(x_i)} t_{(\beta/2, m-n)} < x_i^* < x_i + \sqrt{var(x_i)} t_{(\beta/2, m-n)}$$

Where  $x_i^*$  is the true solution. For these calculations, a 95% confidence interval was used, meaning 2.5% is tolerated on each side (Van Dongeren, 1994).

### F-test

For each flowrate observed, data collected at the same radial and axial positions, but different angular positions, are considered to be “replicates”. Therefore, each data set for a given column, packing, and bed depth yields eight replicate sets of six measurements, if two angular positions are used. A first test of model adequacy is to perform an F-test to show adequacy of fit. The pure-error sum of squares is calculated by:

$$SSPE = \sum_1^N \left( \sum_1^6 \sum_1^8 (T_{observed} - T_{average})^2 \right)$$

Where N is the number of bed depths. Indices are omitted for clarity and averages are taken over each set of eight replicate measurements for each position. The mean square pure error is then calculated by:

$$MSPE = \frac{SSPE}{n_2}$$

Where  $n_2 = m - (\text{number of averages found})$ . The mean square pure error is an estimate of the error variance. Since this varies weakly over the bed depths, it must be estimated by averaging the error variance over the bed-depths for the “overall” analysis case. The mean lack-of-fit sum of squares is then calculated by:

$$MSLF = \frac{S - SSPE}{n_1}$$

Where  $n_1 = m - \text{number of parameters estimated} - n_2$ . If the model is linear in the parameters, the mean lack-of-fit sum of squares is an independent estimator of error variance. However, if the model is nonlinear, then it is a biased estimator, and a test of model inadequacy may be stated as:

$$F_c = MSLF / MSPE > F_{0.05}(n_1, n_2)$$

With values of  $F_c > F_{0.05}$ , either the model is a poor fit or the data has a low amount of variability. This indicated a lack of fit error is significant compared to inherent scatter in the data.

## Results

In order to proceed *with* any results, it was necessary to first prove that the data collected was representative of what was expected compared to past experiments. This includes the following:

- Plots of  $\theta$  vs.  $y$  should show a parabolic fit at the first bed depth
- On the Plots of  $\theta$  vs.  $y$  for each Reynolds number, temperatures should be lower with a higher bed depth
- On a height to height basis,  $k_r/k_f$  and  $Nu_w$  should be height independent
- Peclet and Biot numbers are in line with previously found literature figures

### Parabolic Fit

Over the vast amounts of experiments done with fixed bed reactor tubes, it has been found that temperature profiles in the tubes fit a parabolic curve of the form  $T = A * r^2 + B$ , or in dimensionless form,  $\theta = A * y^2 + B$ . Figure 7 shows an example of a parabolic fit for the first depth for raschig rings in the 4 inch column. As expected, the data follows a parabolic fit very closely with a coefficient of determination of 0.99. It would also be expected that  $\theta_{y=0} = 1$  for the best fit, this is due to how the  $\theta$  is calculated in terms of the center temperature and as so would only hold true if the center of the column had not cooled more than the surrounding locations. This example has an intercept (A) of 1.005, very close to 1. Some temperature profiles for other packings are not as parabolic as expected. Figure 8 is an example of a parabolic fit for the first height of JM 4-hole cylinders. This poor parabolic fit was uncommon however and only occurred in two instances, the raschig rings and the JM 4-hole cylinders in the 2 in column, both of which were instances with a particularly low N value. Graphs expressing the parabolic fit for first depth temperature profiles for each packing/column combination can be found in Appendix B:  $\theta$  vs.  $y$  for First Heights.



# First Height Parabolic Fit for Raschig Rings

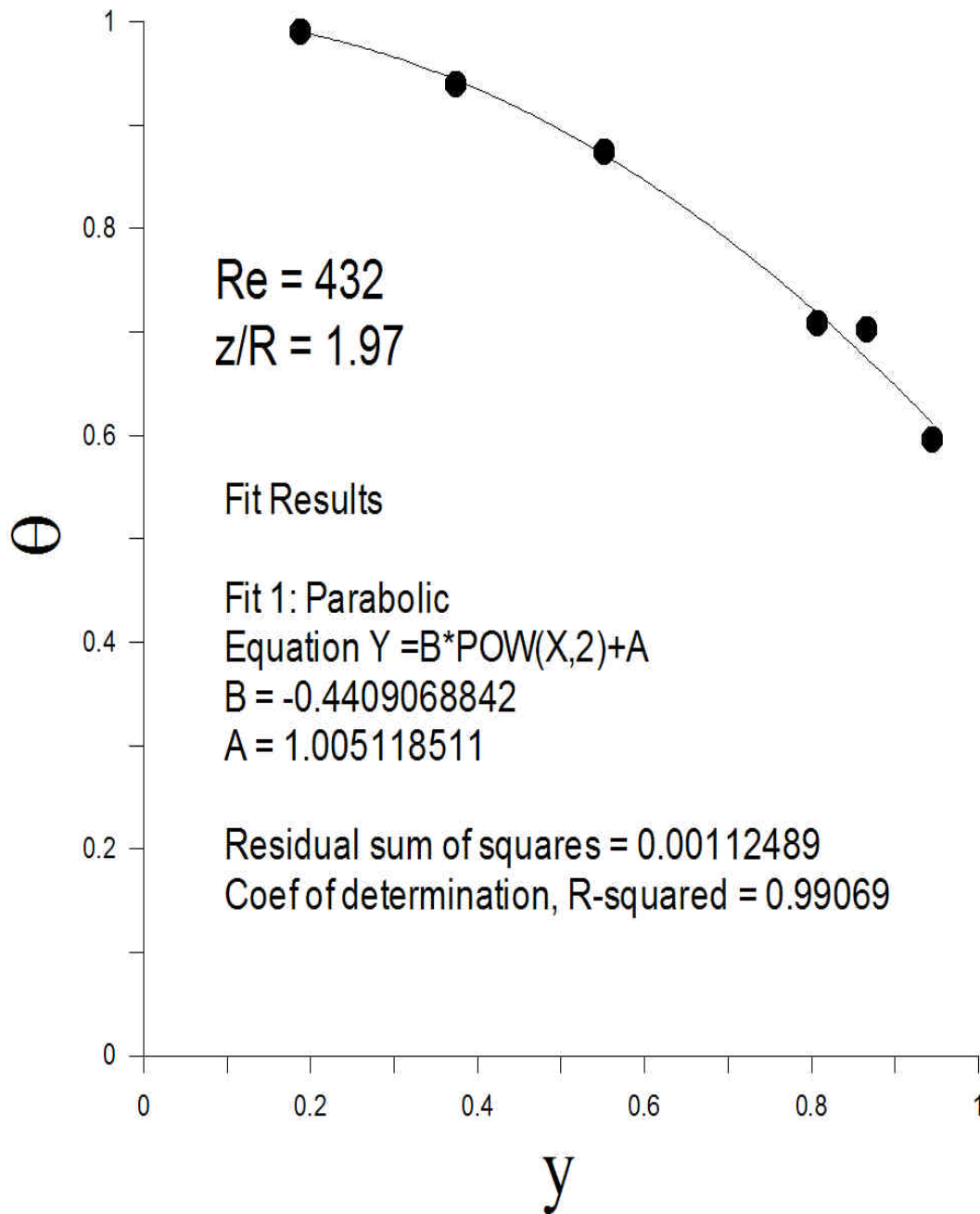


Figure 7: Temperature Profile Parabolic Fit for Raschig Rings in the 4 inch column with Reynolds Number 432

## First Height Parabolic Fit for 4-hole cylinders

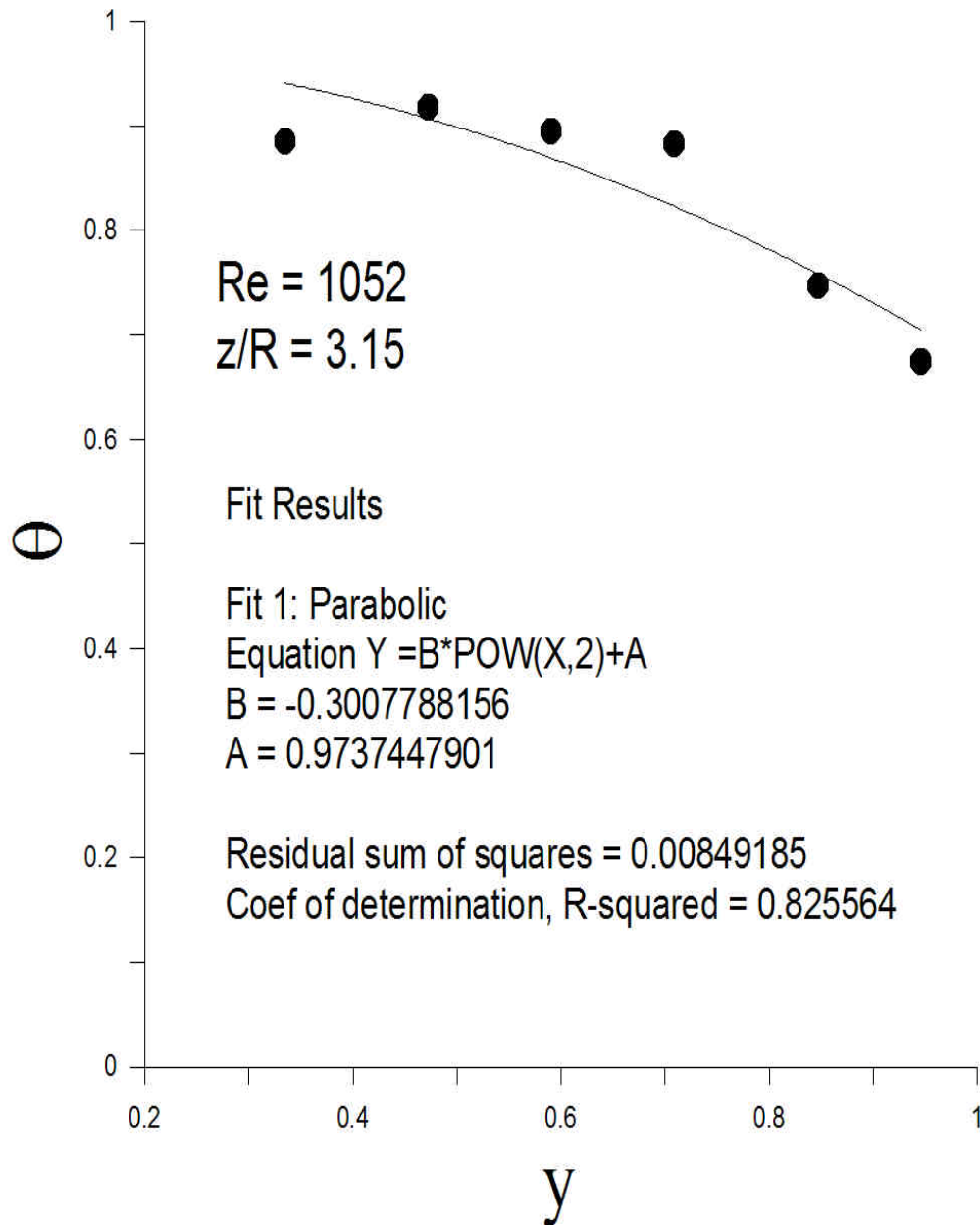


Figure 8: Poor parabolic fit for temperature profile of JM 4-hole cylinders in the 2 inch column with Reynolds Number 1052

The dimensionless profiles should also make physical sense. Since this is a cooling experiment, higher bed depths should yield lower temperatures. Temperatures should also decrease farther from the center of the tubes. It would also make sense for temperature differences between successive bed depths to be lower at greater bed depths. As air travels through the tube, the temperature difference between the center of the tube and the walls is decreased, lowering the driving force of the heat transfer. This means that the rate of temperature decrease with increasing radial position should also decrease with increasing bed depths. A sample radial temperature profile is shown in Figure 9.

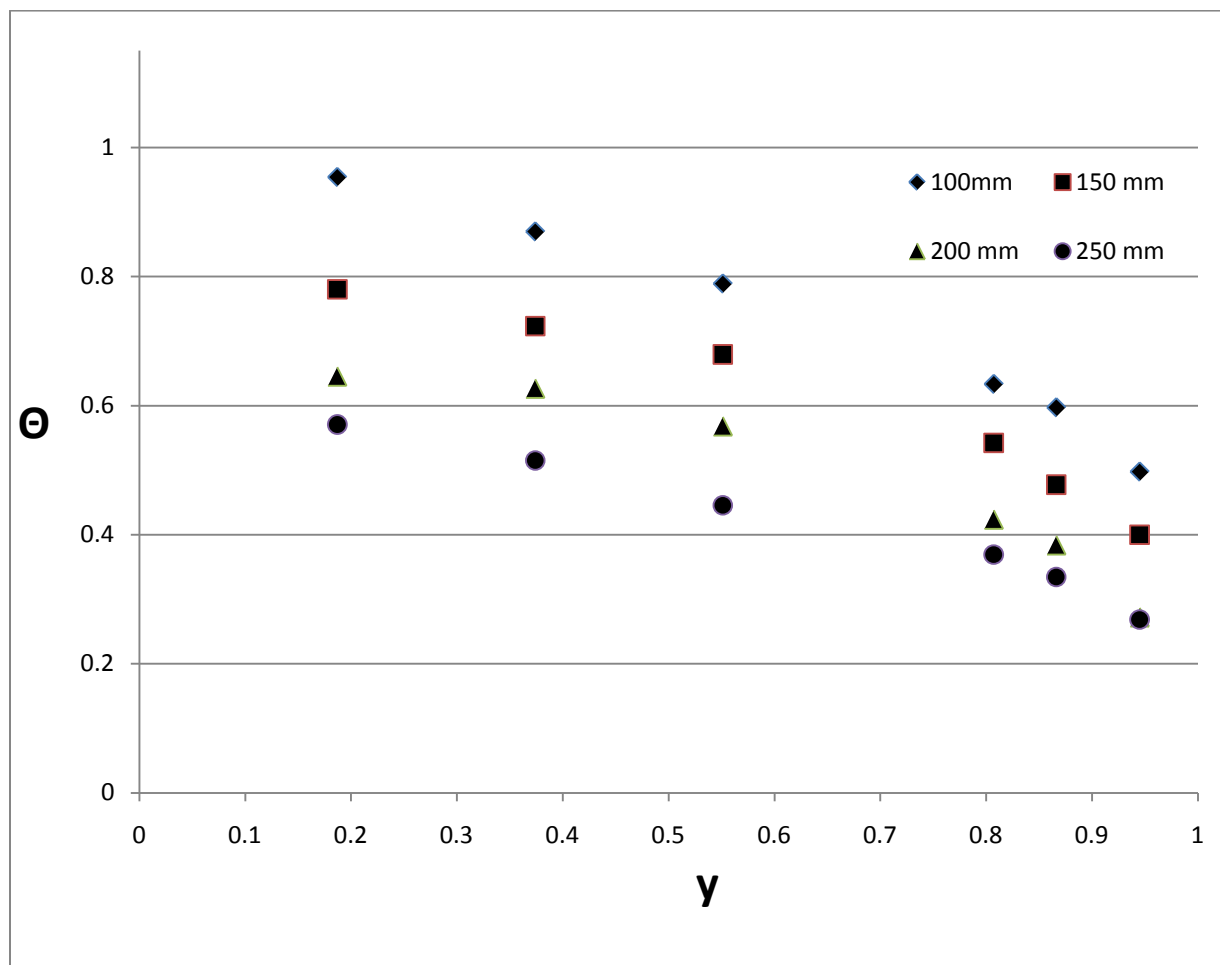


Figure 9: Temperature Profile for JM 4-hole cylinders in the 4 inch column for Reynolds Number 268

Figure 9 is representative of the majority of our temperature profiles, and shows the expected trends for radial profiles obtained during the experiment. It would also be expected for higher

Reynolds numbers to yield higher temperatures. Figure 10 is a temperature profile of the same packing/column as Figure 9 but with a higher Reynolds number.

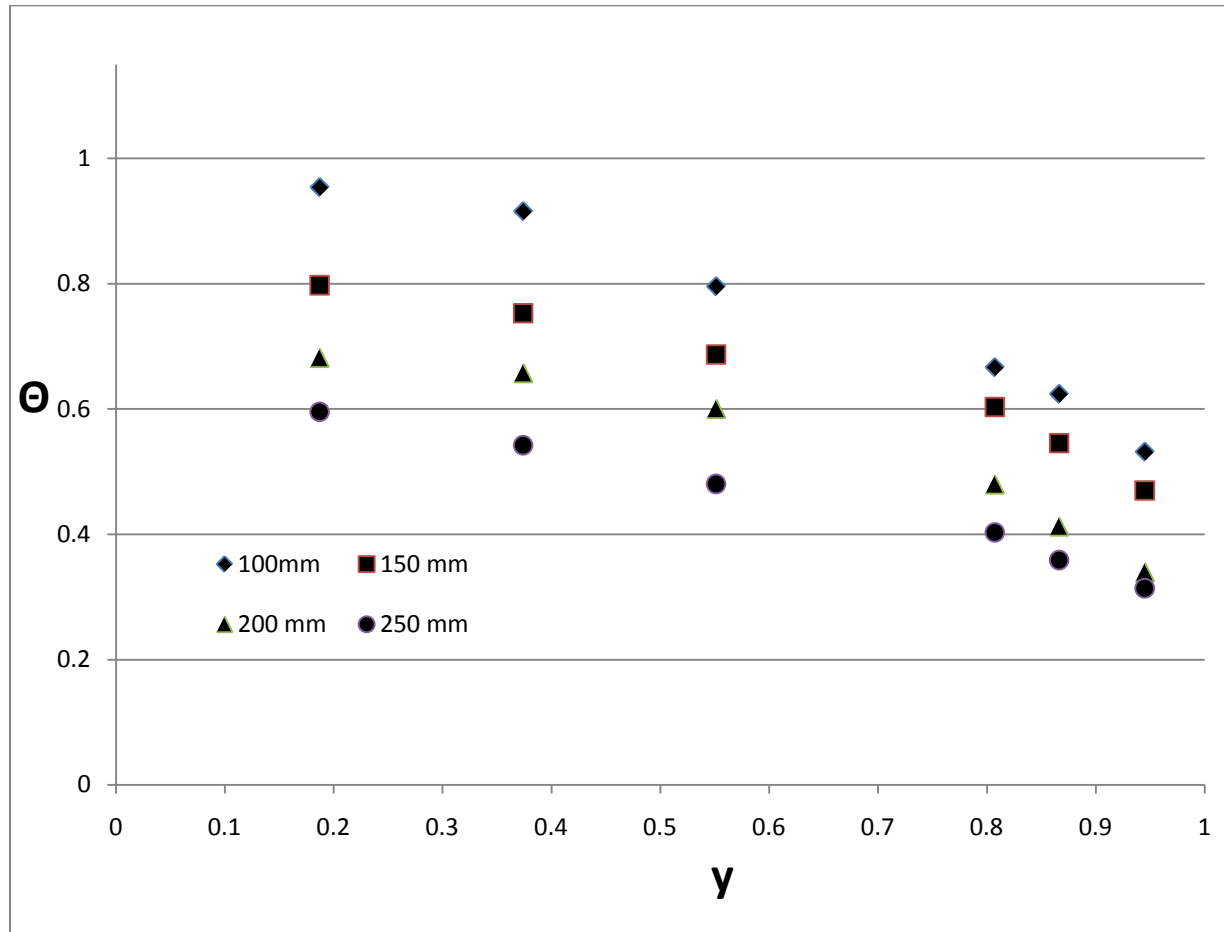


Figure 10: Temperature Profile for JM 4-hole cylinders in the 4 inch column for Reynolds Number 358

Each data point on Figure 10 corresponding with the same bed depth and radius as points on Figure 9 have higher temperatures. Figure 11 displays a temperature profile for a given bed depth rather than Reynolds number.

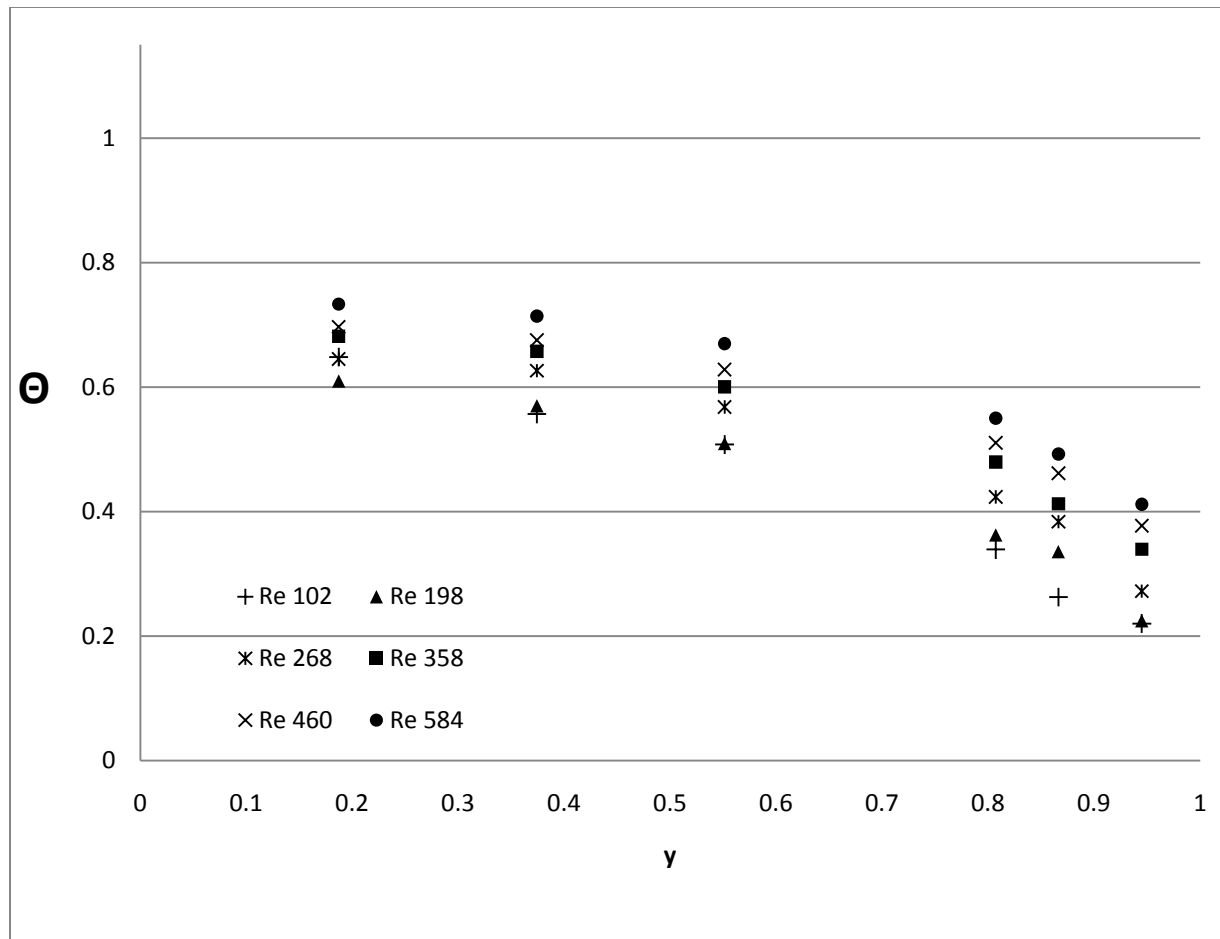


Figure 11: Temperature Profile for JM 4-hole cylinders in the 4 inch column for Bed Depth 200mm

There were few temperature profiles that did not represent our expected findings. Some of the temperatures for higher bed depths would be higher than those for lower bed depths, as well as some temperatures at higher radii being higher than those closer to the center of the tube. This can be a result of bent thermocouples where even a slight deviation from their expected position would cause such a discrepancy. Furthermore since most of the uncharacteristic behavior was seen in the near wall region it could be the result of wall effects dominating this portion of the tube. Figure 12 is an example of these findings.

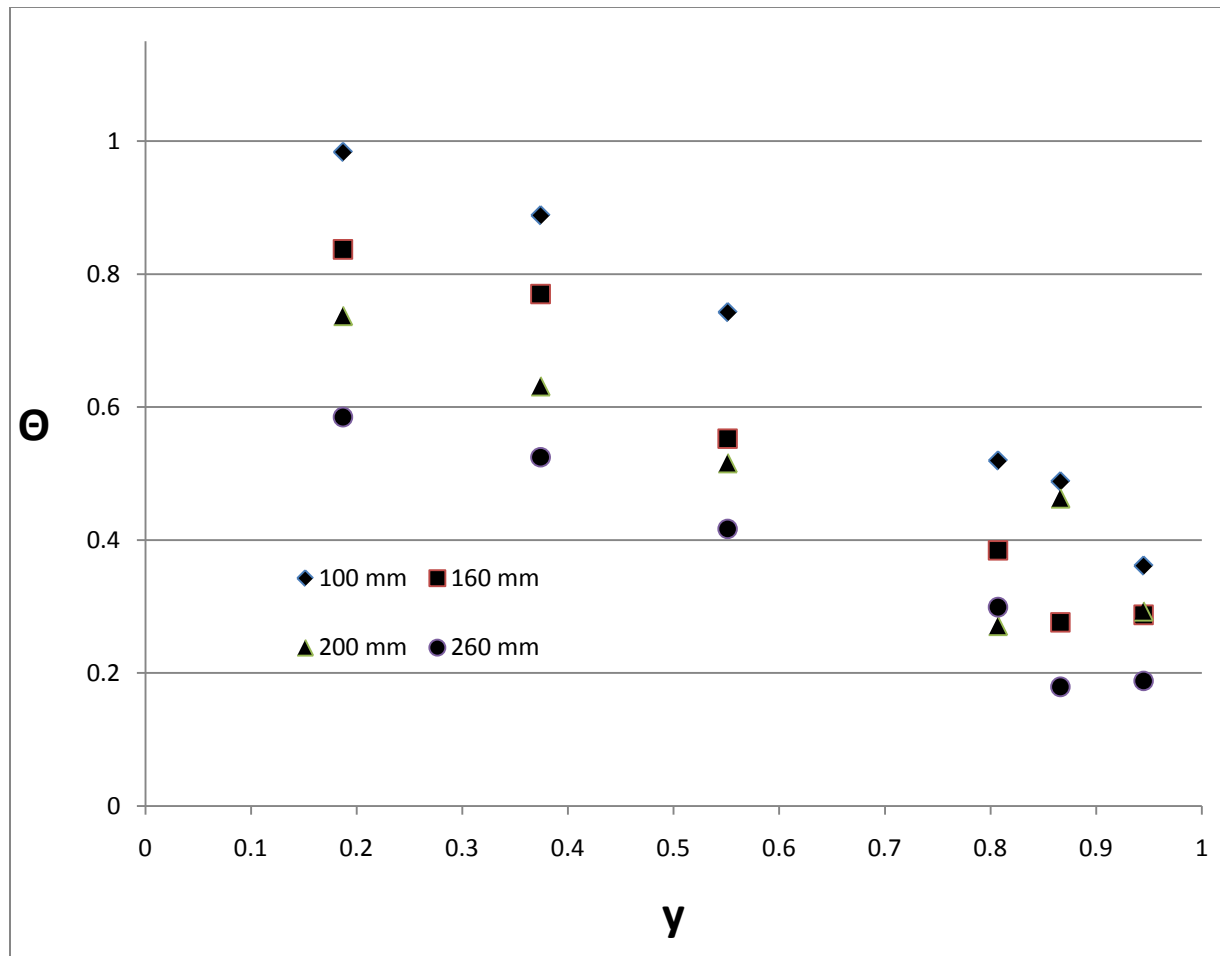


Figure 12: Temperature profile for Monoliths in the 4 inch column with Reynolds Number 129

It is also understood that  $\theta$  should not exceed 1, as  $T_o$  should be the greatest temperature recorded; however, for some packings,  $\theta$  values of greater than 1 were recorded. This also contradicts prior statements regarding the fit being parabolic with an intercept of 1. Larger than expected  $\theta$  values were only recorded for raschig rings in the 2 inch column ( $N=3.5$ ). An example of this is shown in Figure 13. This could be the result of the larger currents created by the larger packing. Because the larger packing takes up a much larger fractional area of the tube cross section it can in effect, if positioned in a certain manner, channel the hot air around the center thermocouple effectively creating a cool spot in this region and resulting in  $\theta$  of over 1.

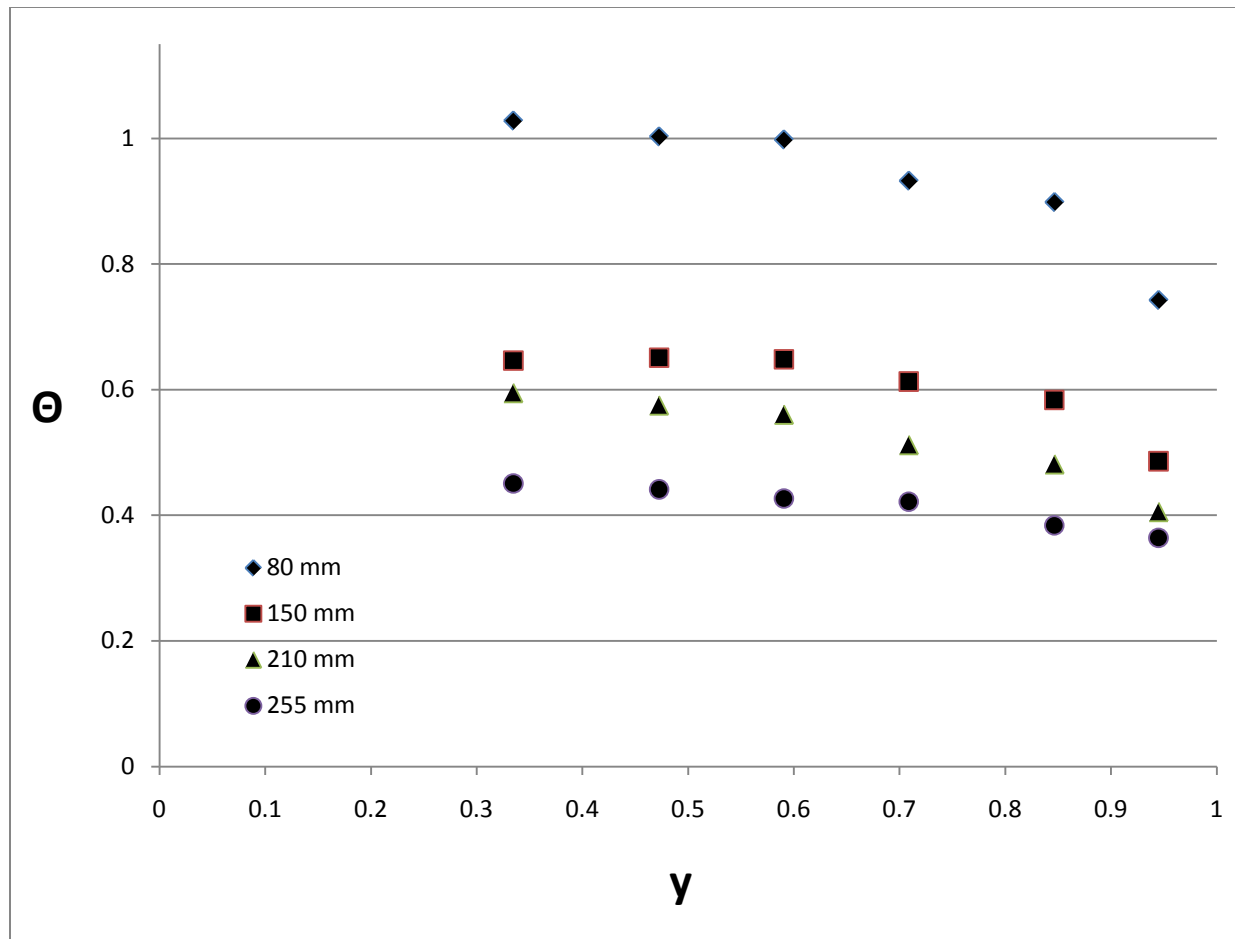


Figure 13: Example of  $\theta$  exceeding 1; Raschig Rings in the 2 inch column with Reynolds number 1570

41 different sets of Reynolds numbers were examined over this course of this project (5 for the spheres, 6 for the other 6 packing/column combinations). Radial temperature profiles for all 41 sets of Reynolds numbers can be found in Appendix C:  $\theta$  vs.  $y$  by Reynolds Number.

### Depth independence on heat transfer parameters

The heat transfer coefficients  $k_f/k_f$  and  $Nu_w$  are strictly based on the packing size and shape, and airflow. It was once assumed that these parameters were both expected to decrease with bed depth because the models used to fit the parameters were not accounting for the axial dispersion of heat (DeWash & Froment, 1972; Gunn & Khalid, 1975). However, it has been shown that bed depth should not be a factor in determining the heat transfer coefficients if the IPPF model is used. In order to show this, the data for the second, third, and fourth heights completed were

compared directly to the first height and values were found for the coefficients. Sample graphs of height by height analysis are shown in Figures 14 and 15, using IPPF model. The vertical error bars for all data collected in this study represent 95% confidence intervals.

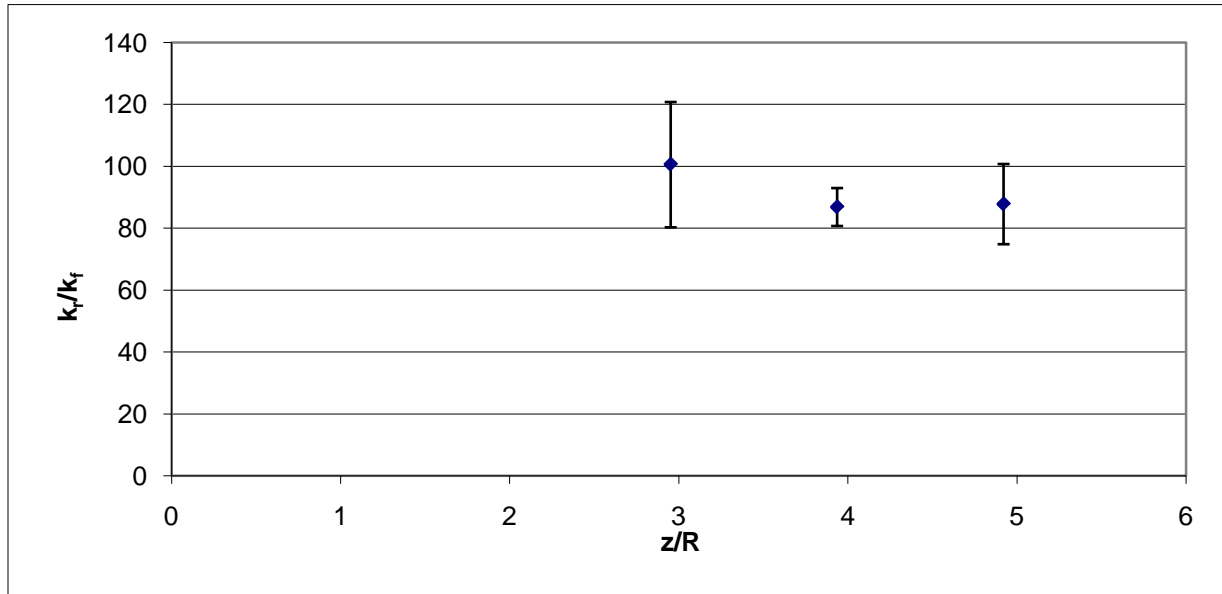


Figure 14: Height by height analysis of  $k_r/k_f$  for JM 4-hole cylinders in the 4 inch column with Reynolds Number 584

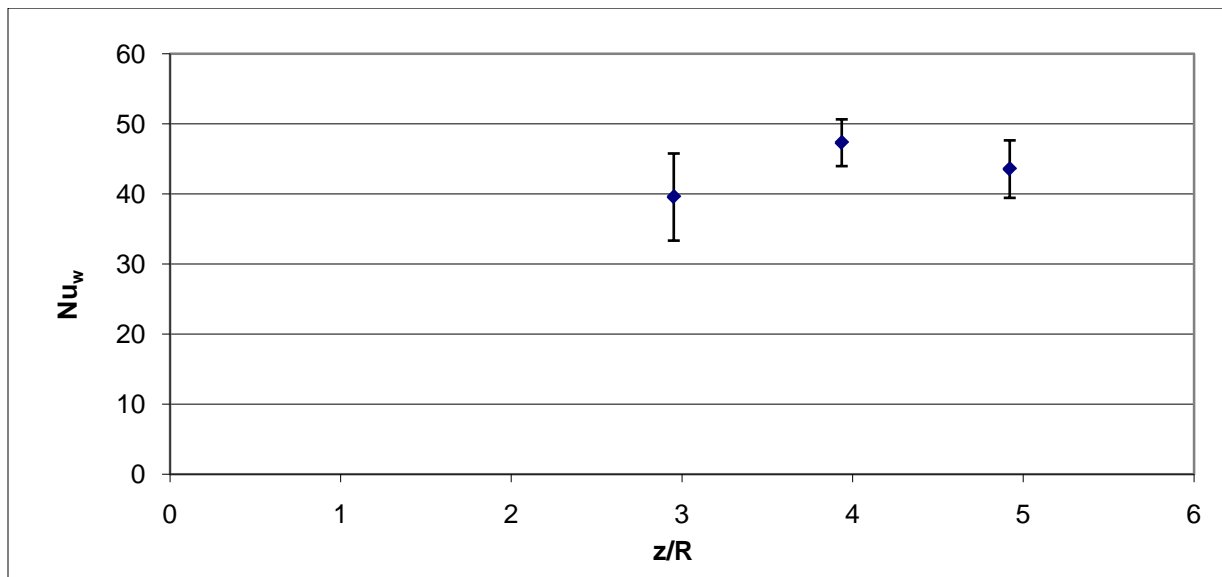


Figure 15: Height by height analysis of  $Nu_w$  for JM 4-hole cylinders in the 4 inch column with Reynolds Number 584



Most data obtained presents no strictly positive or negative trends. For respective bed depths, if  $k_r/k_f$  decreased,  $Nu_w$  would usually increase, and vice versa. Since the overall heat transfer coefficient,  $U$ , is dependent on both of these, increasing one and decreasing the other will not affect this parameter. Figure 16 represents how  $h_w$  and  $k_r/k_f$  are representative of this, showing that using the following equation for the overall heat transfer coefficient:

$$\frac{1}{U} = \frac{1}{h_w} + \frac{\alpha R}{k_r}$$

An increase in  $Nu_w$  and decrease in  $k_r/k_f$  will yield the same value for  $U$ . This has also been seen in the literature. Landon (1996) also showed this; this is shown in Figure 16.

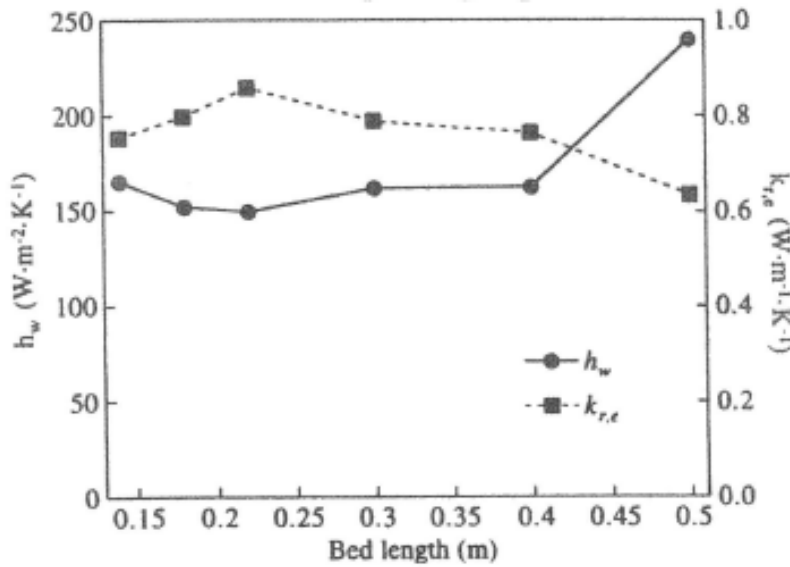


Figure 16: Illustration of bed depth dependence for heat transfer parameters (Landon, 1996)

## Peclet and Biot number analysis

Graphs of the Peclet and Biot numbers were also compared to expectations. All except for one, the graphs are representative of our expectations. As stated before, it was expected according to their definitions that the Peclet number would increase rapidly at low Reynolds numbers and

level out at a certain value of the Reynolds number. Figure 17 presents the Peclet number over a range of Reynolds numbers for 4-hole cylinders in the 4 inch column.

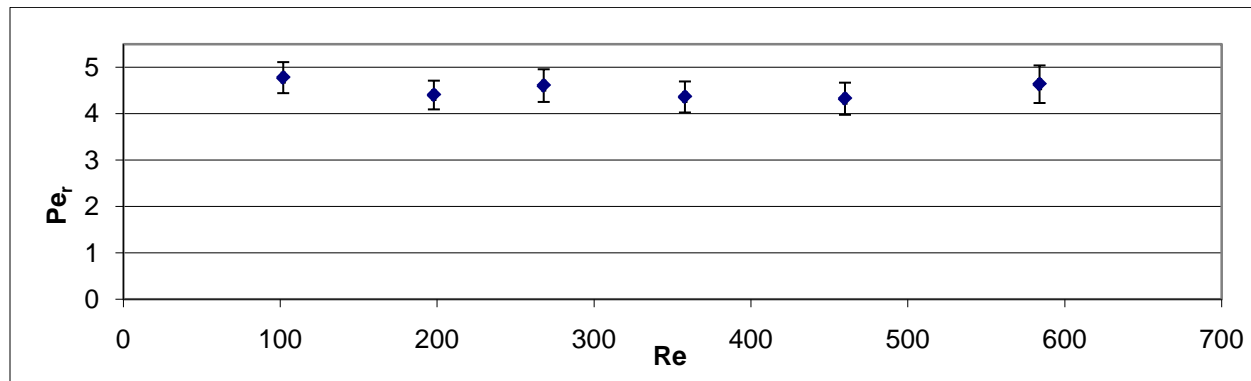


Figure 17: Peclet number versus Reynolds Number for JM 4-hole cylinders in the 4 inch column

The data presented above shows that the Peclet number does not deviate with respect to Reynolds number. The data also shows that the Peclet number for Monoliths is approximately 4.5, which is believable compared to the expected 6 for ceramic particles. This lower value indicates superior heat transfer and as such would make the particle more appealing. Since lower Reynolds numbers were found not to yield reliable data, the values at which Peclet increases rapidly with respect to Reynolds number were not examined. One of the packing/column combinations yielded data unrepresentative of our expectations. This is shown in Figure 18.

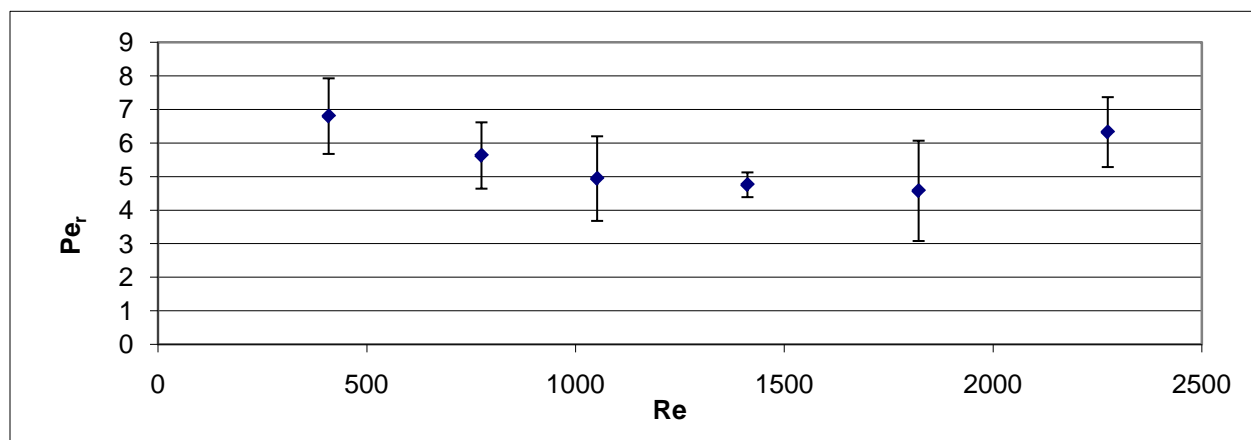


Figure 18: Peclet Number versus Reynolds Number for JM 4-hole cylinders in the 2 inch column

It is very unlikely that  $Pe_r$  would behave in such a manner, and the reasoning for such behavior is uncertain. This might be due to the small N value not allowing the heat to disperse evenly across the cross section of the column, causing a discrepancy in parameters at different flowrates.

The Biot number is expected to decrease with respect to Reynolds numbers. All data of Biot number versus Reynolds number obtained in these experiments expressed this behavior. A sample of these graphs is shown in Figure 19.

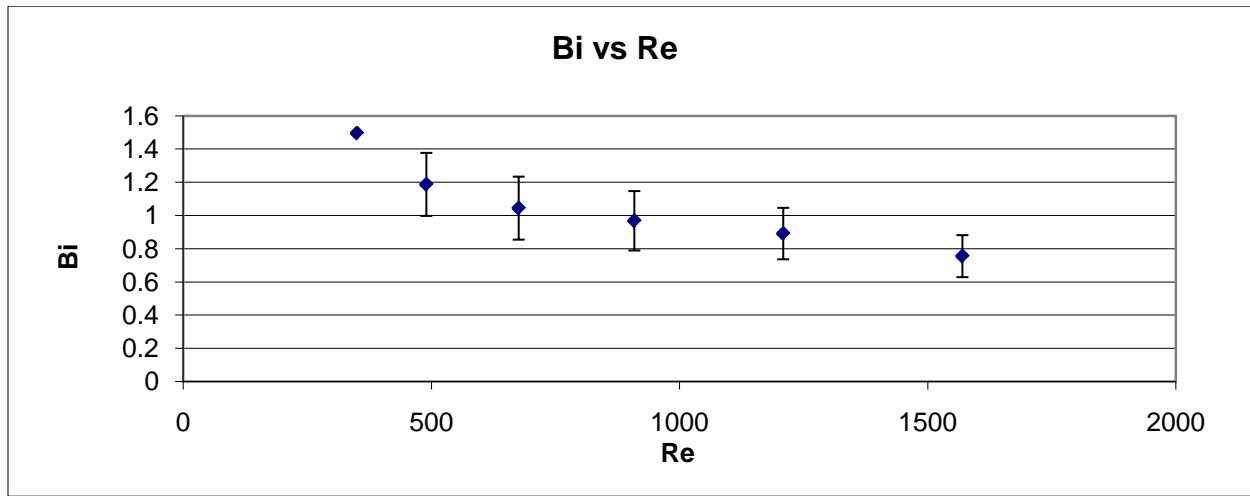


Figure 19: Biot Number versus Reynolds Number for Raschig Rings in the 2 inch column

Most values for the Biot number fall in the range of 1-2, which is close to the expected value. All graphs of Peclet and Biot numbers plotted against Reynolds numbers can be found in Appendix E:  $k_r/k_f$ ,  $Nu_w$ ,  $Bi$ , and  $Pe_r$  for all Packings and Columns.

### The Bauer/Schlunder Method

A study done by Smirnov (2002) showed an equation for finding  $k_r/k_f$ :

$$k_r/k_f = k_r/k_{f,bed} + K * Re * Pr$$

Where  $Pr$  is the Prandtl number for air, which is assumed constant at 0.71. The parameter  $k_r/k_{f,bed}$  is the effective radial thermal conductivity of the packing with stagnant air. This parameter must be determined experimentally by plotting  $k_r/k_f$  versus Reynolds number and finding the y intercept of the best fit line. In Smirnov's (2002) study, this was assumed to be 10 for ceramic particles (regardless of shape or size) and 20 for glass particles. Although this is much less significant than the slope of the line (considering steam methane reforming is undergone at very high Reynolds numbers), it is important to verify that this value is close to expected to ensure that the slope is accurate. This means that having a negative  $k_r/k_{f,bed}$  value will yield a slope that cannot be assumed to be perfectly accurate, and therefore extrapolating will not be precise.

This leaves  $K$ , which may be determined experimentally or mathematically through the Bauer/Schlunder method.  $K$  varies for each packing, and depends on size ( $N$ ), shape, and material. Experimentally, this would be the slope of the  $k_r/k_f$  versus  $Re$  plot divided by the Prandtl number. The Bauer/Schlunder method (Bauer & Schlunder, 1978) for determining  $K$  can be expressed by:

$$K_{Bauer} = \frac{X}{8 * \left\{ 2 - \left[ 1 - \left( \frac{1}{R/d_p} \right) \right]^2 \right\} * d_p}$$

Where  $X$  for cylindrical particles can be expressed as:

$$X = \frac{\varepsilon_{bed}}{\varepsilon_{hbed}} F_{cyl} d_p + \frac{\varepsilon_{hole} (1 - \varepsilon_{bed})}{\varepsilon_{hbed}} F_{hole} d_{hole}$$

Where  $F_{cyl} = 1.75$ ,  $F_{hole} = 2.8$ ,  $d_{hole} = \sqrt{2}l$ ,  $\varepsilon_{bed}$  can be assumed as 0.40 for cylindrical particles,  $\varepsilon_{hole}$  is the porosity of one particle, and  $\varepsilon_{hbed} = \varepsilon_{bed} + \varepsilon_{hole} (1 - \varepsilon_{bed})$ . For Spherical particles,  $X$  can be expressed as:

$$X = 1.15d_p$$

Experimental K values were compared with those found mathematically using the aforementioned equations. The Bauer/Schlunder equation does not correlate our results well. However, correcting the equation to read:

$$K_{Bauer,corrected} = \frac{X}{7 * \left\{ 2 - \left[ 1 - \left( \frac{1}{R/d_p} \right) \right]^2 \right\} * d_p}$$

By correcting the  $K_{Bauer}$  equation, it was found that the equation correlates our experimentally found K values nicely. This yields the following K values:

**Table 6: Comparison of  $K_{Bauer}$  and  $K_{exp}$  to account for corrected equation**

Packing	$K_{Bauer}$	$K_{Bauer,corrected}$	$K_{exp}$	N
Raschig Ring (4 inch)	0.226	0.258	0.252	7.0
JM 4-hole cylinders (4 inch)	0.195	0.223	0.225	5.8
Raschig Ring (2 inch)	0.185	0.212	0.175	3.5
JM 4-hole cylinders (2 inch)	0.161	0.184	0.183	2.9

The value of 8 used in the denominator of the  $K_{Bauer}$  equation has been found for an ideal model. Since the experiment was not ideal, changing this value to 7 works similar to that of a correction factor to correct for the reality of this experiment.

A study completed by Borkink et al. (1993) compared Peclet number (Note that this is not the radial Peclet number, but instead  $Pe = Re * Pr$ ) to the effective radial thermal conductivity for different packings including raschig rings with an equivalent spherical diameter of 6.2mm in a 49.9mm column (N=8). This was compared to data obtained in this study for raschig rings in the 4 inch column (N=7). To obtain the Peclet number, the Reynolds number was multiplied by the Prandtl number for air. This comparison is displayed in Figure 20.

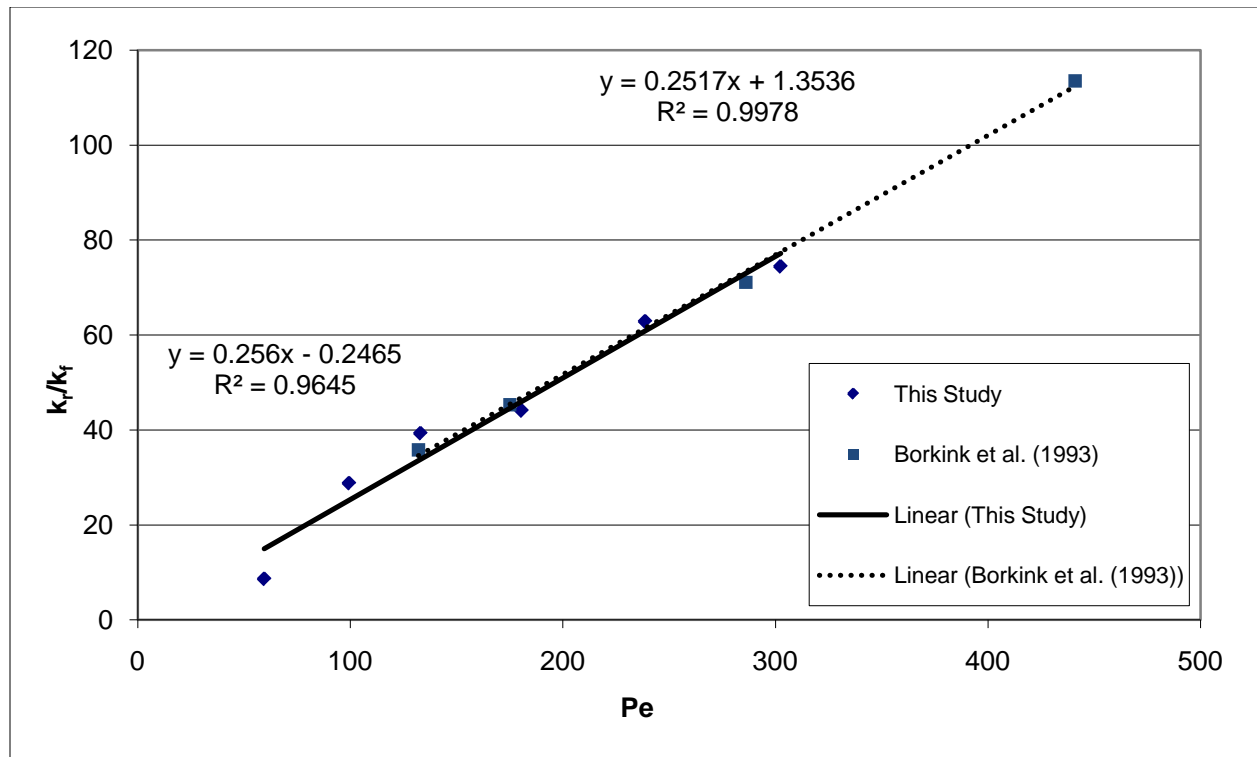


Figure 20: Comparison of Borkink Raschig Ring effective radial thermal conductivity to values from this study

The slopes for both best fit lines are nearly identical, verifying that the value determined for  $K$  (which is the slope for the best fit lines above) using the corrected Bauer and Schlunder equation is accurate (according to this and Borkink et al.).

### Comparison to past experiments using spheres

The data from this experiment was also compared to various other studies which used ½ inch porous ceramic sphere packing in the 4 inch column. These studies were performed by Hans Van Dongeren (1994) and Darryl Pollica (Dixon, Personal Communication). The two studies were both performed using a heating method rather than cooling; cool air entered the column at the inlet and steam in the jacket heated the air up. Although these were not cooling experiments as this study was, the two are comparable as the IPPF model adjusts all variables to be dimensionless.

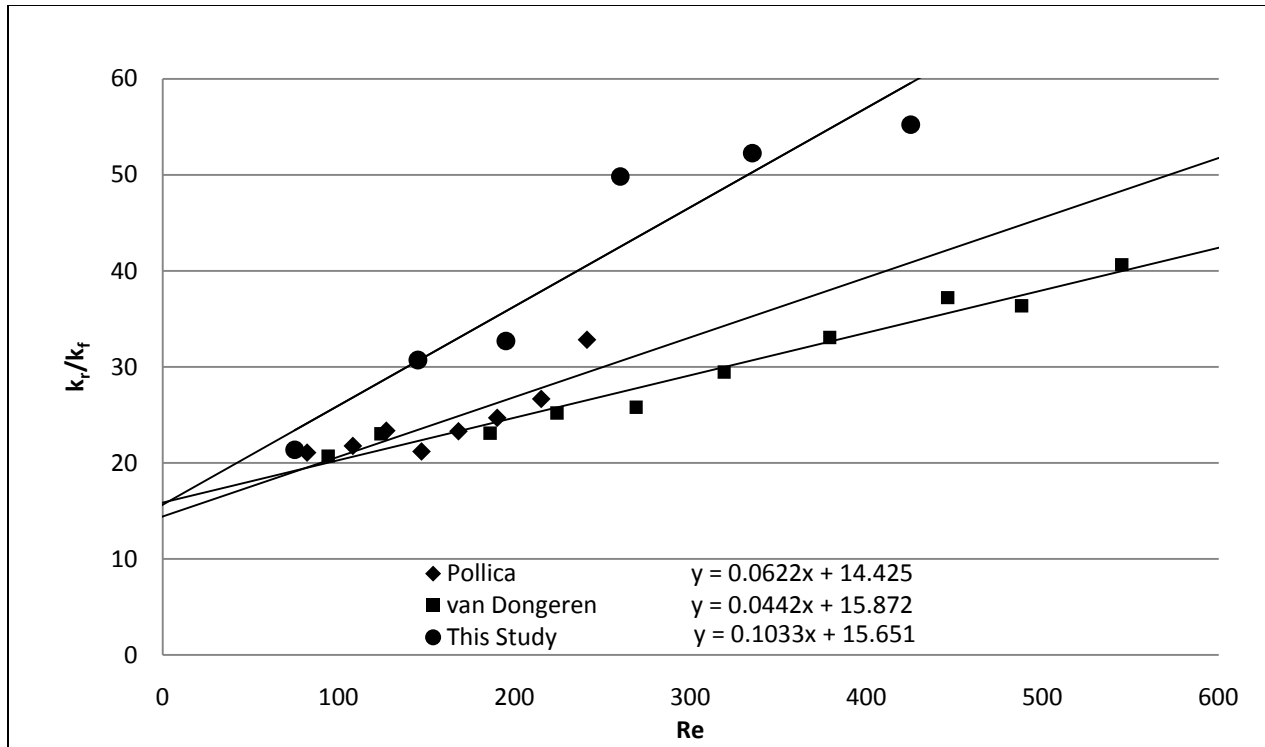


Figure 21: Comparison of  $k_r/k_f$  for spheres

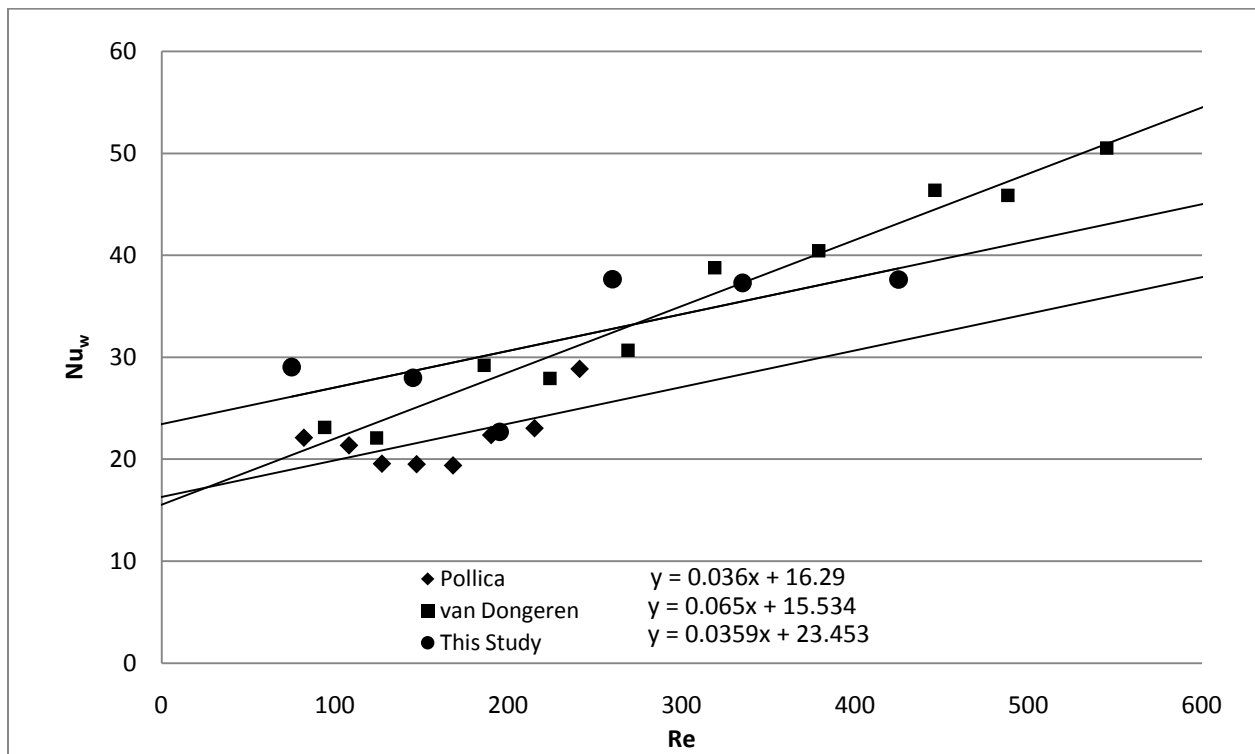


Figure 22: Comparison of  $Nu_w$  for spheres

The  $k_r/k_f$  comparison illustrates that this study, Pollica, and van Dongeren all determined that the intercept should lie somewhere around 15, however, none of the slopes strongly correlate from one study to another. Although it appears that van Dongeren has a stronger correlation with more data points than the other two studies, a slope (where slope =  $K \cdot Pr$ ) of 0.044 is far less than the expected 0.07 for a spherical packing with  $N=8$  (Bauer & Schlunder, 1978).

Although  $Nu_w$  is not linear in nature with respect to Reynolds number, it can be compared using linear trends at low flowrates. The intercept collected in this study deviates greatly from the intercepts obtained by Pollica and van Dongeren (which are both about 16). However, disregarding the first data point (at a Reynolds number less than 100), the intercept is less than 20. The slope in this study is identical to that of Pollica, which shows that both our study and Pollica's will yield similar  $Nu_w$  values at very high flowrates.

### Asymptotic radial Peclet relationship to $k_r/k_f$

Von Landon (1996) stated that at high Reynolds numbers, one may disregard  $k_r/k_{f,bed}$  in calculating  $k_r/k_f$  by using the equation:

$$k_r/k_f = \frac{Re * Pr}{Pe_{r,f}^{\infty}}$$

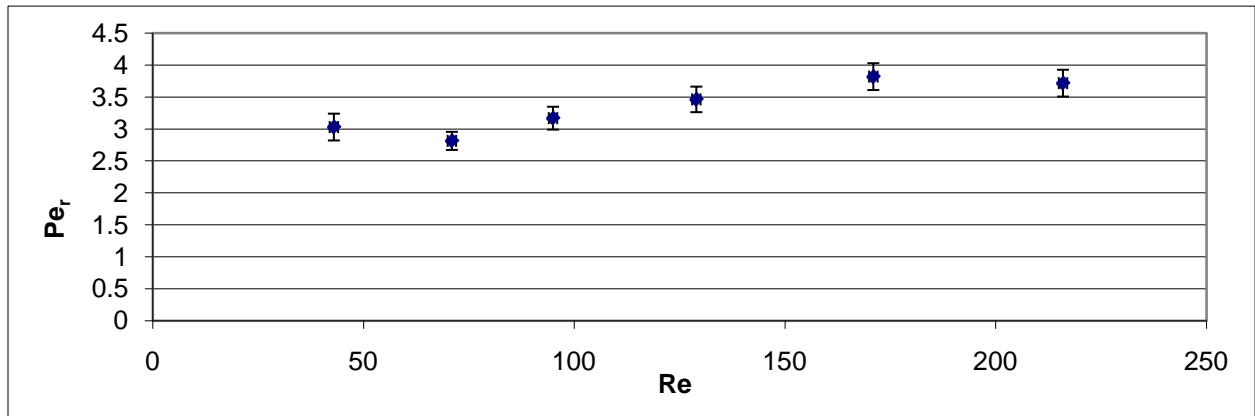
Where  $Pe_{r,f}^{\infty}$  is the radial asymptotic Peclet number at high Reynolds numbers. This equation states that  $Pe_{r,f}^{\infty} = \frac{1}{K}$ . There are empirically derived equations for  $Pe_{r,f}^{\infty}$ , such as Bauer and Schlunder, but when using different shaped particles, these equations cannot be assumed accurate. To determine this value, these experiments need to be run at high Reynolds numbers. However, with varied data for  $Pe_r$  at higher Reynolds numbers, this calculation cannot be assumed perfect. With the data taken in this study,  $Pe_{r,f}^{\infty}$  is the approximate average of  $Pe_r$  values over the last 4 Reynolds numbers in which data was recorded. For example, for 4-hole cylinders in the 4 inch column (Figure 17), values for  $Pe_r$  average 4.5.



**Table 7: Correlation between experimentally found slopes and radial Peclet numbers**

Packing	$K_{exp}$	$Pe_{r,f}^{\infty}$	$1/Pe_{r,f}^{\infty}$	N
Raschig Ring (4 inch)	0.252	4	0.25	7.0
JM 4-hole cylinders (4 inch)	0.225	4.5	0.22	5.8
Monoliths (4 inch)	0.225	3.5	0.29	14.0
Raschig Ring (2 inch)	0.175	6	0.17	3.5
JM 4-hole cylinders (2 inch)	0.183	6	0.17	2.9
Monoliths (2 inch)	0.225	4.5	0.22	7.0

The only packing that greatly deviates from  $K_{exp}$  is the monoliths in the 4 inch column. The correlation between Peclet and Reynolds numbers is shown in Figure 23.



**Figure 23: Peclet versus Reynolds number for monoliths in the 4 inch column**

As shown in Figure 23, the Peclet number is increasing over Reynolds numbers instead of remaining constant. This is most likely due to the flowrates being too low for the Peclet number to asymptote, and therefore  $Pe_{r,f}^{\infty}$  is not definable for this packing. According to the experimental K value determined by the slope,  $Pe_{r,f}^{\infty}$  should be 4.5, and very well might be. Comparing  $1/Pe_{r,f}^{\infty}$  and slopes of  $k_r/k_f$  to Reynolds number is also useful to confirm that the data has been collected at high enough flowrates where the flow is fully developed. The lower the value for  $Pe_{r,f}^{\infty}$ , the higher  $k_r/k_f$  is, meaning that the heat transfer would be overestimated for cases such as this.

## Analytically determining a Nusselt wall value

Martin and Nilles (1993) proposed that  $Nu_w$  may be determined analytically by using the following equation:

$$Nu_w = \left(1.3 + \frac{5}{N}\right) k_r / k_{f,bed} + 0.19 Re^{0.75} Pr^{0.33}$$

In a simpler form, this equation is:

$$Nu_w = A + 0.1697 Re^{0.75}$$

Where A is the intercept based on the packing's N and  $k_r/k_{f,bed}$  value. There are equations to determine theoretical values for  $k_r/k_{f,bed}$ , however, this study used the values of the intercepts from graphs of  $k_r/k_f$  versus Reynolds number. Since Prandtl is assumed constant for air at 60°C to be 0.71, the second term is solely dependent on Reynolds number. The data collected in this study does not support the accuracy of this equation. An example of this equation compared to this study is shown in Figure 24.

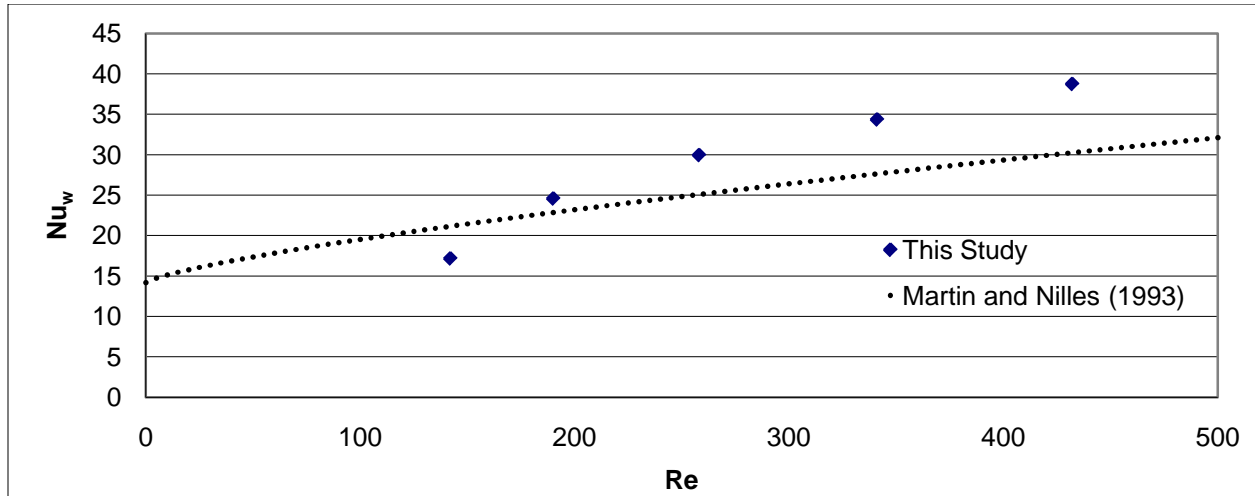


Figure 24:  $Nu_w$  versus Re for raschig rings in the 4 inch column

In order to determine correct values for  $Nu_w$ , the intercept of  $k_r/k_f$  versus Re graphs must be accurate. Some of the data in this study yielded intercepts far lower than expected (some negative) which causes the Martin and Nilles comparison to be far off from the collected  $Nu_w$  data. The Martin and Nilles equation compared to this study's data for 4-hole cylinders in the 4 inch column is shown in Figure 25.

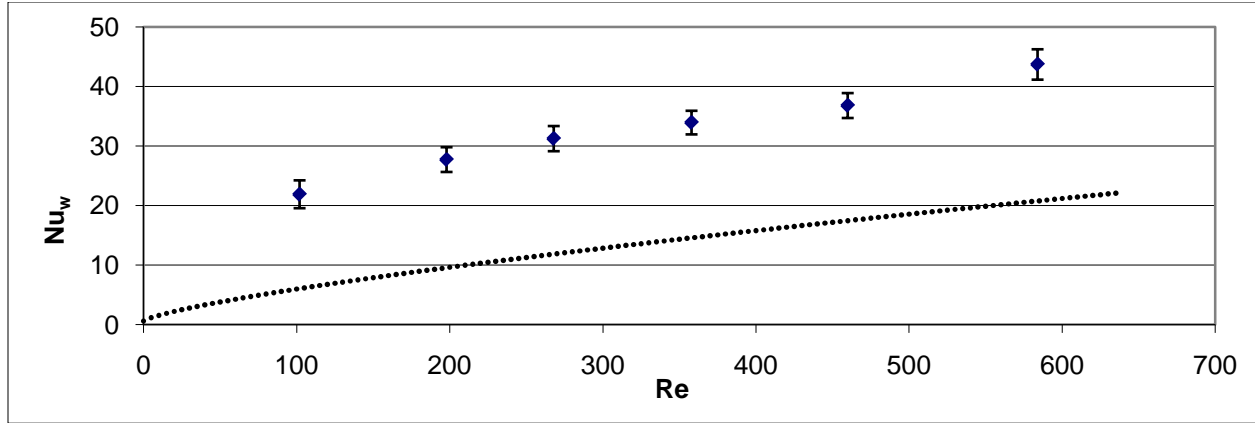


Figure 25:  $Nu_w$  versus  $Re$  for 4-hole cylinders in the 4 inch column

It can be seen that the series created by the Martin and Nilles is relatively parallel to the data obtained in this study; this deviation of the equation to collected data is solely dependent on the intercept ( $k_r/k_{f,bed}$ ). After correcting the  $k_r/k_{f,bed}$  value to be 8 instead of the obtained 0.2835 for the 4-hole cylinders in the 4 inch column, a much better correlation was found. This is shown in Figure 26.

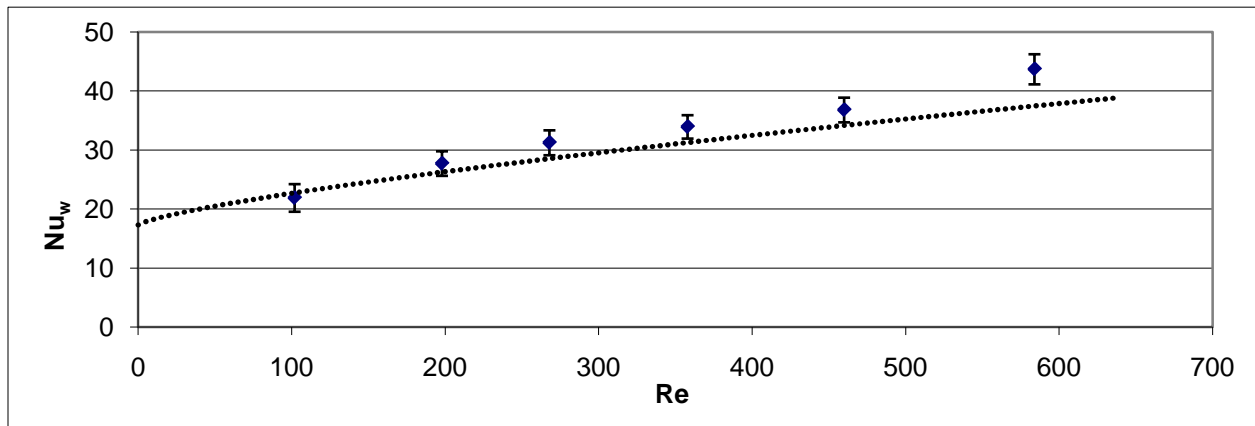


Figure 26:  $Nu_w$  versus  $Re$  for 4-hole cylinders in the 4 inch column with corrected  $k_r/k_f$  bed

## N Dependence

Through the course of the experiment the value of  $N$  ranged from 2.9 to 14 depending on the packing and tube, the full list can be found in Table 2. Typically  $N$  can be expected to fall in the range of 5 to 15 (Von Landon, 1996) and it is generally accepted that this range will produce the desired heat dispersion profiles. As the  $N$  drops significantly lower than this range you start to encounter data that does not fit the predicted profiles. This is largely due to the wall effects which dominate the region within a particle diameter of the tube wall (Smirnov, 2002).

Therefore, for any  $N \leq 2$ , the entire tube region would be affected entirely by wall effects;  $N$  must be large enough so that the majority of thermocouple readings are not affected by this phenomena. This can be observed by looking at the parabolic fits for the runs where the Raschig Rings and the JM 4-hole cylinders in the 2in column,  $N=3.5$  and  $N=2.9$  respectively (figures 5 and 6), by far produced the least parabolic curves at the first bed heights. Further evidence of poor model prediction in this region comes from values of  $\theta$  larger than one found for the Raschig Rings in the 2 inch column as well as an uncharacteristic Peclet versus Reynolds correlation for the JM 4-hole cylinders in the 2 inch column. These findings come in contrast to a study done by Dixon (1997) which concluded that an  $N$  of as low as 2 for non-sphere particles will still produce the desired results.

### Particle Durability

All the particles in this study were composed of ceramics however they varied in both shape and size. It was found that the monolith packing, being of smallest size and having the thinnest walls was far more brittle than the other packings. While the packing heights in this study remained below 300mm in all instances it was still found that many monoliths at the bottom of the column, whether as a result of weight or agitation cracked and crumbled. The raschig rings were far more durable than the monoliths however even these resulted in a few broken particles most likely during the initial packing of the tower. The JM 4-hole cylinders and the spheres were by far the most resilient of the particles as none of these showed cracking or breaking.

### Analysis of Variance

In order to ensure that the data is accurate, the F-ratio of  $F/F_{95}$  should be less than 1. However, it was found that for packings in the two inch column, values larger than 1 were very common. It was also found that values larger than 1 were often found for monoliths in the 4 inch column. For the raschig rings and 4-hole cylinders in the 2 inch column, this was most likely due to the low  $N$  value, causing less uniform heat dispersion, causing the sum of squares to be very large. However, since this is not the case with the monoliths, the discrepancy may be attributed to the large voidage within the particle. A table with all F-ratio values can be found in Appendix F: F-test values.

## Conclusion

The IPPF model requires that the lowest height of packing used should provide a  $z/R$  ratio of 3. It has been stated that a value below this results in insufficient development of the profile at the inlet height resulting in failure of the model. However, data collected in this study for  $z/R$  ratios of 2 showed parabolic profiles. In future studies, if this is found not to be the case, a new model would need to be utilized that does not require a parabolic fit of the temperature profile.

The value of  $N$  displays a positive correlation to  $Nu_w$  resulting in a distinctive rise in the latter when  $N$  was double by moving to the smaller tube. Furthermore an  $N \leq 3.5$  is dominated by wall effects and results in data that does not fit the model while  $N \geq 5.8$  fits the model well. Further studies would need to be conducted in the range of  $3.5 \leq N \leq 5.8$  to determine at which point the wall effects start to dominate a sufficiently large portion of the tube to negate the model.

Analyzing our data for the packings in the 4 inch column, we were able to not only get nearly identical findings for raschig rings, but were able to determine analytical models for determining heat transfer parameters based on shape and size. The Bauer/Schlunder equation, when corrected, gave parameters identical to those collected experimentally. It was also found that Von Landon's  $Pe_{r,f}^{\infty}$  correlation to  $k_r/k_f$  is a fair estimation. It was however found that the Martin and Nilles equation for determining  $Nu_w$  cannot be relied on for accurate measurements as it only depends weakly on particle shape.

The JM 4-hole cylinders and Monoliths studied provided the best heat transfer parameters. However, since the Monoliths are brittle and would be subjected to high forces during steam methane formation, these should not be considered for industrial processes. Further studies should compare the differences between similar shapes such as 3-hole, 4-hole, and 5-hole cylinders.

## Nomenclature

Bi	Biot Number ( $Nu_w \frac{R/d_p}{k_r/k_f}$ )
$Bo_{h,r}^\theta$	Bodenstein Number ( $\frac{G C_p R^2}{k_r L}$ )
$C_p$	Heat capacity
$d_p$	Particle diameter
$d_{hole}$	Characteristic length of the particle ( $\sqrt{2}l$ )
$d_t$	Tube diameter
G	Mass flowrate of the gas
$h_w$	Wall heat transfer coefficient
$k_r$	Effective radial thermal conductivity
$k_f$	Effective thermal conductivity of the fluid
F	Variance
L	Largest bed length analyzed
N	Tube to particle diameter ratio ( $d_t/d_p$ )
$Nu_w$	Wall Nusselt number ( $\frac{h_w d_p}{k_f}$ )
$Pe_r$	Radial Peclet number ( $\frac{Re * Pr}{k_r/k_f}$ )
$Pe_{r,f}^\infty$	Asymptotic radial Peclet number ( $\frac{Re * Pr}{k_r/k_f}$ at high Re values)
Pr	Prandtl number (0.71 for air at 60 °C)
r	Radial position
R	Radius of the tube
Re	Reynolds number ( $\frac{v_s \rho d_p}{\mu}$ )
T	Temperature
$T_o$	Center temperature of the first bed depth
$T_w$	Wall temperature
U	Overall heat transfer coefficient
$v_s$	Superficial velocity
x	Dimensionless bed depth ( $z/R$ )
y	Dimensionless radial position ( $r/R$ )
z	Bed depth
$\alpha$	Experimentally determined constant
$\varepsilon_{bed}$	Bed porosity without account of porosity of particle
$\varepsilon_{hbed}$	Overall bed porosity
$\varepsilon_{hole}$	Porosity of one particle
$\lambda$	Effective radial thermal conductivity (same as k)
$\mu$	Viscosity
$\rho$	Density
$\theta$	Dimensionless temperature ( $\frac{(T-T_w)}{(T_o-T_w)}$ )
$\zeta$	Dimensionless bed depth ( $\frac{x^R/L - x_o^R/L}{1 - x_o^R/L}$ )

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## Appendices

## **Appendix A: Sample Fortran Input Text File (for JM 4-hole cylinders in the 2 inch column)**

4	6	3	2		
50.8	17.4244				
8.5	12	15	18	21.5	24
2275	80	0			
98.6993515					
73.51792602		78.67514038		77.5713257	77.86281738
77.3210266		80.45686036		76.06216734	73.99614106
76.99973906		74.60718994		76.78083646	77.72229768
69.05369872		79.96601562		77.00187682	68.09851226
69.2140457		69.6364151		69.71127474	71.04101106
63.0244606		60.36585998		72.53876342	60.9682289
19.60186308		21.4395725		25.94641496	
2275	80	45			
98.77065276					
73.27800448		75.00024414		79.92648164	76.18695678
73.44379732		77.1693451		75.37802274	78.39521024
70.07826082		76.28060302		78.61668702	74.13243866
69.04063262		73.66384276		73.69291838	71.69318848
72.88054352		68.1328476		71.98683624	72.6760376
63.12102432		61.88073728		69.87362978	65.75786592
19.51357994		21.3685955		25.99892502	
1822	80	0			
99.0032837					
72.00116424		73.03620758		78.05301664	74.74094236
71.51310426		75.45122678		74.21852876	77.48835146
67.20661316		74.16232908		78.2910904	73.26902616
69.28578644		71.16259612		73.7652176	71.61493072
70.35703428		71.04037018		68.31296846	69.6669754
60.4158493		60.79260866		72.35227966	65.32909394
20.29633062		22.00180892		25.93361932	
1822	80	45			
98.91923066					
77.16016844		70.76894226		73.54588778	75.07192536
68.4898941		74.84447022		77.00720824	76.84577026
79.58320314		72.13587802		74.36732786	74.35321196
76.23908386		61.07149504		69.64187774	76.7695099
72.09566194		70.35791016		62.07328492	72.56248322
50.56270676		64.38652344		57.6177574	60.06700898
20.44216992		22.02308922		25.89328118	
1412	80	0			
98.76869048					
73.5621277		67.0405426		70.00103302	71.15197754
65.63481446		71.73593292		73.14741362	73.5072876
76.6353195		68.17412874		70.96861876	70.14902648
72.67462006		57.41319732		66.3239807	73.62355348
67.15747682		66.8013733		58.54669038	68.66848298
47.83555144		60.07205964		53.94326248	57.07460404
19.71891554		21.07039492		24.4406624	
1412	80	45			
98.9111679					
73.34187928		69.24677278		72.00898896	69.62782288
67.89277344		73.18645324		75.1600876	70.95996706
70.12002104		71.0735321		71.23355406	70.35523376
74.25852206		65.93112794		71.36910858	65.40884704
64.26127778		60.37468032		71.2966324	61.95517502
52.64456712		64.01000594		64.45240936	51.4372383

19.72838284	21.09444772	24.4977024	
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64.68190612	68.25741732	69.82284546	70.71866304
63.92250366	67.90347598	68.50434264	68.65388334
69.14316408	63.49986954	65.06321718	68.72596892
62.48444292	56.15174638	67.8101944	52.43913954
51.44942398	64.33299254	53.92048034	54.6673523
20.2851219	21.36883354	24.19305116	
1052 80	45		
98.89687198			
59.91440584	58.86440276	59.0906105	59.24870224
64.17294388	56.07076494	62.93479616	58.85851134
60.84555434	54.73794404	60.27732468	63.80253372
53.72100146	63.26296768	59.98911668	55.78862226
47.97101974	58.24943086	58.64743574	59.78501664
48.59130326	50.051342	56.23343276	48.8460846
20.27505568	21.15976448	23.92082404	
775 80	0		
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65.95184784	61.06864318	65.21930846	58.41373674
59.70376434	57.51419678	64.41526184	63.91295698
59.09545746	63.09750598	58.552787	53.92599182
51.70502546	61.2921379	56.91301496	58.56207198
47.50985186	55.72848282	53.45356826	44.60525436
20.31828842	21.1653122	23.46459046	
775 80	45		
99.5616104			
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62.53291472	65.90697784	61.09473192	63.43332518
64.0926132	64.39998626	61.02958984	65.32199096
57.55845868	64.65750732	60.02675782	64.38395692
60.25372468	59.1067772	62.8616829	59.52434004
56.02996826	50.4035042	55.21190646	48.15922166
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409 80	0		
118.6779892			
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52.38668366	55.49029696	50.82851026	55.43695144
55.15008468	53.41965256	50.87927324	55.7397484
48.3984268	52.73624648	48.4462166	50.59808884
46.3868744	48.52955476	51.60263214	46.51319352
45.83642578	37.79503402	43.69383314	32.8810745
20.18028944	20.63690606	22.03336868	
409 80	45		
115.8672944			
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32.8019684	38.80463332	42.5204437	30.83472978
20.17482872	20.52550964	21.59088936	
2275 150	0		

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58.43430328	60.83964308	63.91230622	63.82107468
61.44636842	61.42918168	64.08067014	59.02149506
55.13606186	59.46472624	65.10252836	62.940213
63.5492874	60.35691376	63.37976916	56.53418884
56.3140785	65.41085968	61.00628664	51.28270644
48.51304932	57.2770996	63.54102782	53.03800506
19.56986694	22.42404098	27.06655008	
2275 150	45		
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57.95550918	61.47342226	65.20809022	64.25086974
55.06790694	58.79736326	63.9519615	62.98031006
59.12059098	57.87317044	64.7995209	61.86237182
59.14152144	64.8930344	59.46840822	52.28660656
53.22789	53.5156502	59.27845002	51.18025818
19.54683762	22.52379266	27.16913606	
1822 150	0		
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50.29889906	56.62985916	60.96076582	58.29543536
47.95373078	51.76853638	58.13508528	58.63936234
56.99634626	49.17751694	60.99326704	58.00704422
54.63617708	60.73489074	55.4197731	47.53644486
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19.2335289	21.498431	25.7419239	
1822 150	45		
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57.09482574	46.32721864	56.52928848	59.86500244
54.8510872	53.31860504	59.7137375	59.45969544
44.55054322	52.48655244	59.7456833	57.80756608
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49.65307238	53.9867485	57.66937942	56.13626404
50.15673296	54.14580612	57.11678008	55.44091338
51.93353118	45.9466507	56.7082733	53.5809471
49.6351883	53.94620438	55.31752014	49.32602538
37.22709578	47.92097246	51.71604768	39.12158202
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1412 150	45		
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38.8451248	47.79931642	49.53569186	47.91036528
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47.18281328	50.41306534	46.15266038	36.77309798
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19.16157456	20.50416106	23.67472992	
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775 150	0		
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46.45537264	49.10773392	49.6659912	45.5958679
49.3028244	45.623275	47.26775512	39.63362196
47.500325	49.26344376	45.51480788	42.73489836
37.46359254	46.86143492	40.74909362	32.50484694
19.61428872	20.74641036	23.3496689	
775 150	45		
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48.47971724	49.90555954	45.859346	46.2270523
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48.4061447	45.473584	43.57394258	46.35172424
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19.61437988	20.76703798	23.37957422	
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31.35536196	37.33917542	29.57197228	33.74812316
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32.75461652	29.23967438	26.69602202	26.30184174
19.19199756	19.6527851	21.40013428	
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37.12656172	36.52278898	30.67872924	34.82304308
31.96548194	37.33473208	29.40726546	34.20025178
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52.09917146	49.93562622	51.71668168	49.73918764
50.47908556	50.70390778	48.12815628	48.78876494
47.19949416	46.8016281	50.37097092	48.32941208
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46.73545458	42.03614576	45.39466628	48.28736572
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40.54960176	39.46253052	43.84951708	48.62294236
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45.2327606	47.04674378	48.03281784	46.26497802
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1052 200	0		
98.90518646			
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42.08356248	44.7333618	42.92609786	41.22968826
38.4459564	41.72648162	44.28848724	40.32664414
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33.66010284	40.84319838	40.1227333	30.39145394
19.6530487	21.6501873	24.25802344	
1052 200	45		
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39.34235308	35.26944732	41.42165374	43.3284668
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775 200	45		
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409 200	45		
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21.42739336	21.35714682	25.24629404	25.95150148
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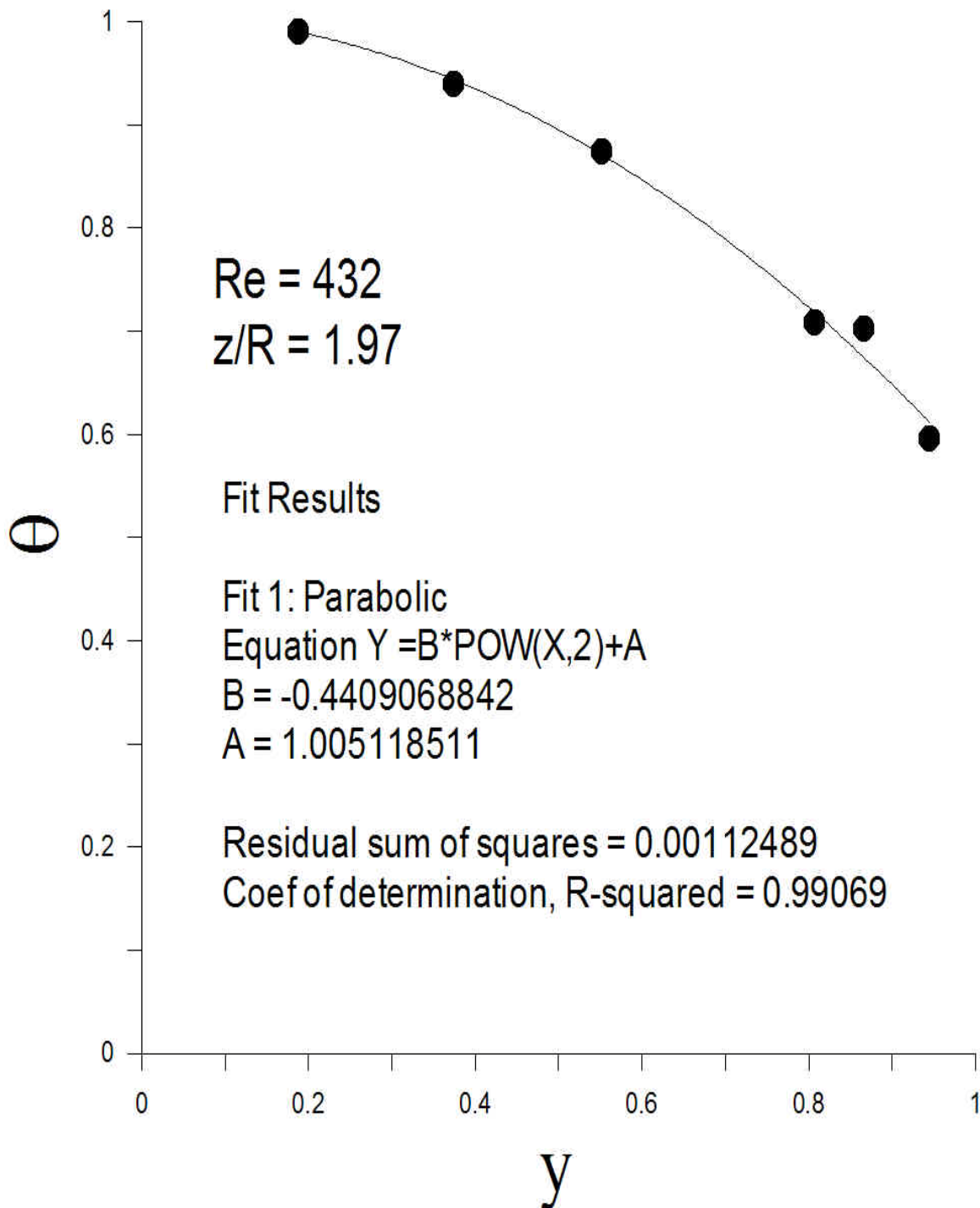
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49.5806618	47.7857712	50.11046524	50.6676811
46.00337676	48.51928712	51.20819552	49.48350982
45.03485108	50.19804764	50.2034294	47.94532014
40.21110228	42.11347884	49.10993194	43.37984618
19.14161796	22.82235376	25.6085289	
1822	265	0	
98.6812439			
45.64598466	45.38593904	46.33173368	46.16474382
47.2903206	43.71267548	48.0881996	45.86076738
45.66292114	44.4619667	46.02790068	47.06360014
41.8697502	45.01825792	47.16648484	45.427948
41.13500598	45.99850846	46.4579643	43.97706374
36.88848192	36.26695482	45.13731002	38.98748628
19.13657722	22.81351854	25.60926932	
1822	265	45	
98.54974824			
40.23674318	41.40471724	41.78658068	43.065905
40.36697006	42.1230606	40.6655464	43.69579698
39.12290878	41.32575532	39.48177186	42.29679184
40.51795654	38.9880913	38.06063768	43.35520782
41.0656563	39.80584792	44.13357772	37.30011594
32.17085114	36.9378525	34.38277056	40.00833436
19.16079788	22.79908218	25.6180031	
1412	265	0	
98.86578064			
38.85801544	40.33667298	41.21251526	39.3657837
41.02693712	39.53769534	42.59932708	38.1136116
38.99116362	39.99701918	41.10022506	41.65617904
38.27532042	40.0788841	42.83440324	37.3191269
37.29112626	40.71051256	40.61609422	37.57031404
33.90524674	35.1454651	41.77336274	31.52881432
19.30854414	22.29714164	24.7952156	
1412	265	45	
98.84623718			
40.84026108	40.28384552	40.93802412	38.18530426
40.0435661	40.25686796	40.24929352	38.56129454
40.50852966	38.9292061	42.20601348	37.72995224
41.57413102	38.55087358	42.70633698	37.83195952
37.91297914	42.59278488	36.45733796	38.74069826
32.4420685	38.94508132	38.72758942	32.00665246
19.34336056	22.30565488	24.81808052	
1052	265	0	
98.09759826			
36.20451658	33.15526428	36.40771636	33.58348694
32.71285326	35.2932205	36.85055848	33.38526384
34.7976776	33.23992082	36.67690506	34.27435608
35.24885482	28.84222374	40.1488388	33.02709502
31.88615686	37.90348968	33.2546242	30.61848222
25.40261496	32.41366576	37.13974838	29.28974496
19.3337341	21.68404466	23.84773864	
1052	265	45	
97.96114348			

35.79308546	32.31847306	35.40328292	35.34646988
35.32076034	32.20355608	38.20213088	34.305616
33.40152894	30.65407182	35.30542906	36.50457916
32.60639648	32.333667	37.0570244	34.15605394
29.6860058	37.62654956	37.27133942	32.78776094
26.0327778	31.92216798	35.86511306	28.8260361
19.38240816	21.70701676	23.86766778	
775 265	0		
99.89899596			
31.82552604	28.38801652	30.5000683	31.62730332
31.06622048	27.57356606	33.3532692	30.88825838
29.44453966	26.32150536	30.79584962	32.26807864
27.96599694	28.06433906	32.06499098	30.6549839
25.32743416	32.47447206	33.15893628	28.53027764
23.201284	26.99083404	30.97368928	26.04771766
19.22278254	20.92047804	22.90697594	
775 265	45		
97.69941254			
31.92369768	28.91629524	31.81234208	30.4450676
27.01779098	30.30033952	31.98719826	31.88331568
28.97728652	27.63662642	34.2344177	30.6048332
32.50058058	26.23759192	31.85536194	29.03033294
27.94880486	31.50609702	30.267054	24.08791198
23.96117554	31.70822486	31.33988838	26.99343032
19.26017456	20.96857226	22.96351282	
409 265	0		
112.4658508			
24.88429414	23.86322706	24.40131724	23.95190508
22.69894904	24.37530212	24.39242288	24.7513981
23.15618592	22.77808152	25.97793732	24.04668348
25.1021969	21.79917872	24.2587055	23.52177584
22.74705086	24.2143673	23.62438622	21.05283624
20.57918664	24.04594078	24.72375946	21.10760954
18.73377798	19.5138748	21.00809098	
409 265	45		
109.804715			
24.10503808	25.07635842	24.41656036	23.32913706
24.43660392	24.4563286	24.6548073	22.89769706
23.58973198	26.3138546	23.8388481	22.91086046
24.27376138	24.76650392	23.57692644	22.27217254
25.59636114	23.14626388	21.51476516	22.94084472
23.4987427	22.99370384	22.18857652	20.10545082
18.7401596	19.53059462	21.00464324	
-1 -1	-1		

## Appendix B: $\theta$ vs. $y$ for First Heights

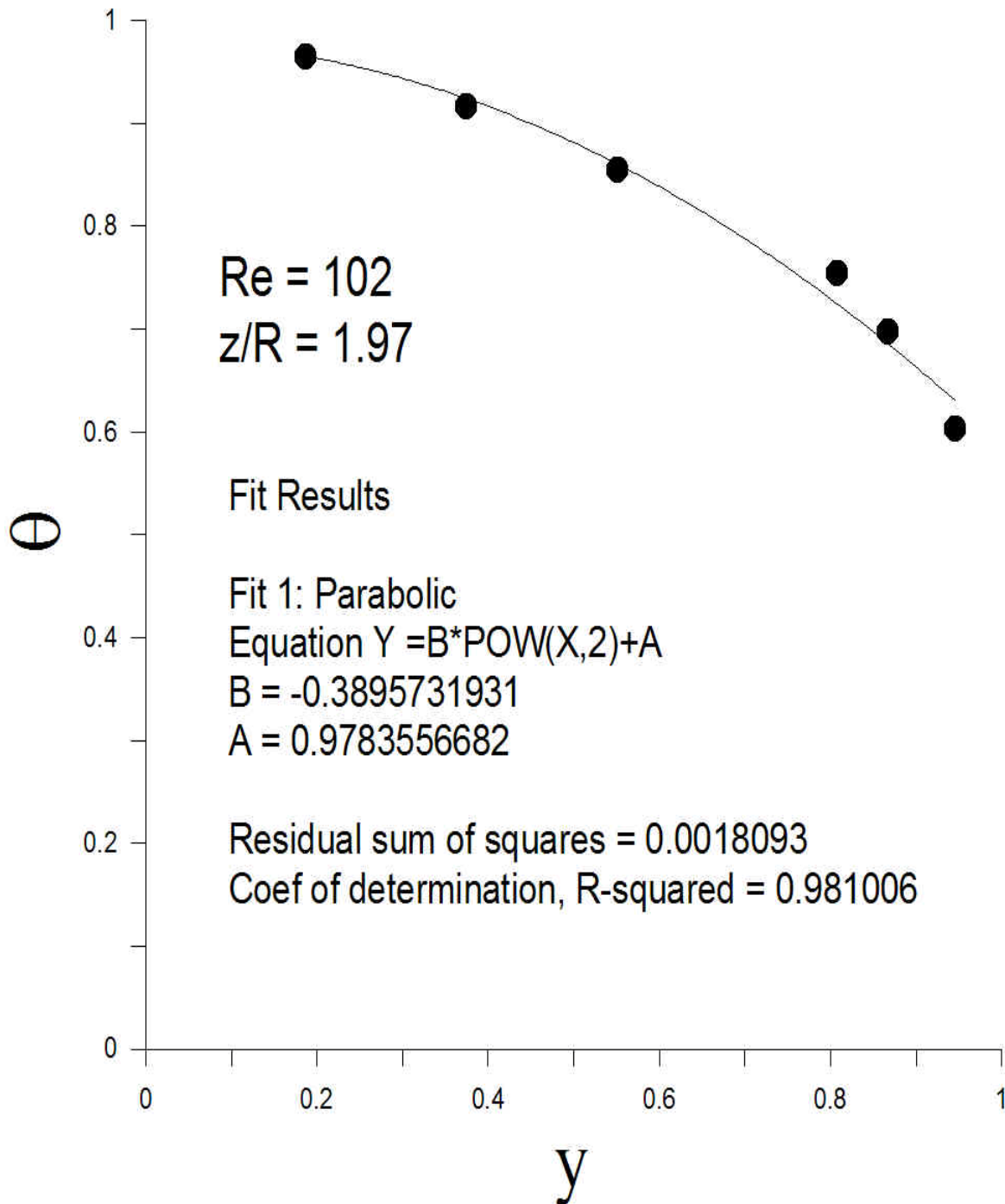
# First Height Parabolic Fit for Raschig Rings

In the 4inch Column



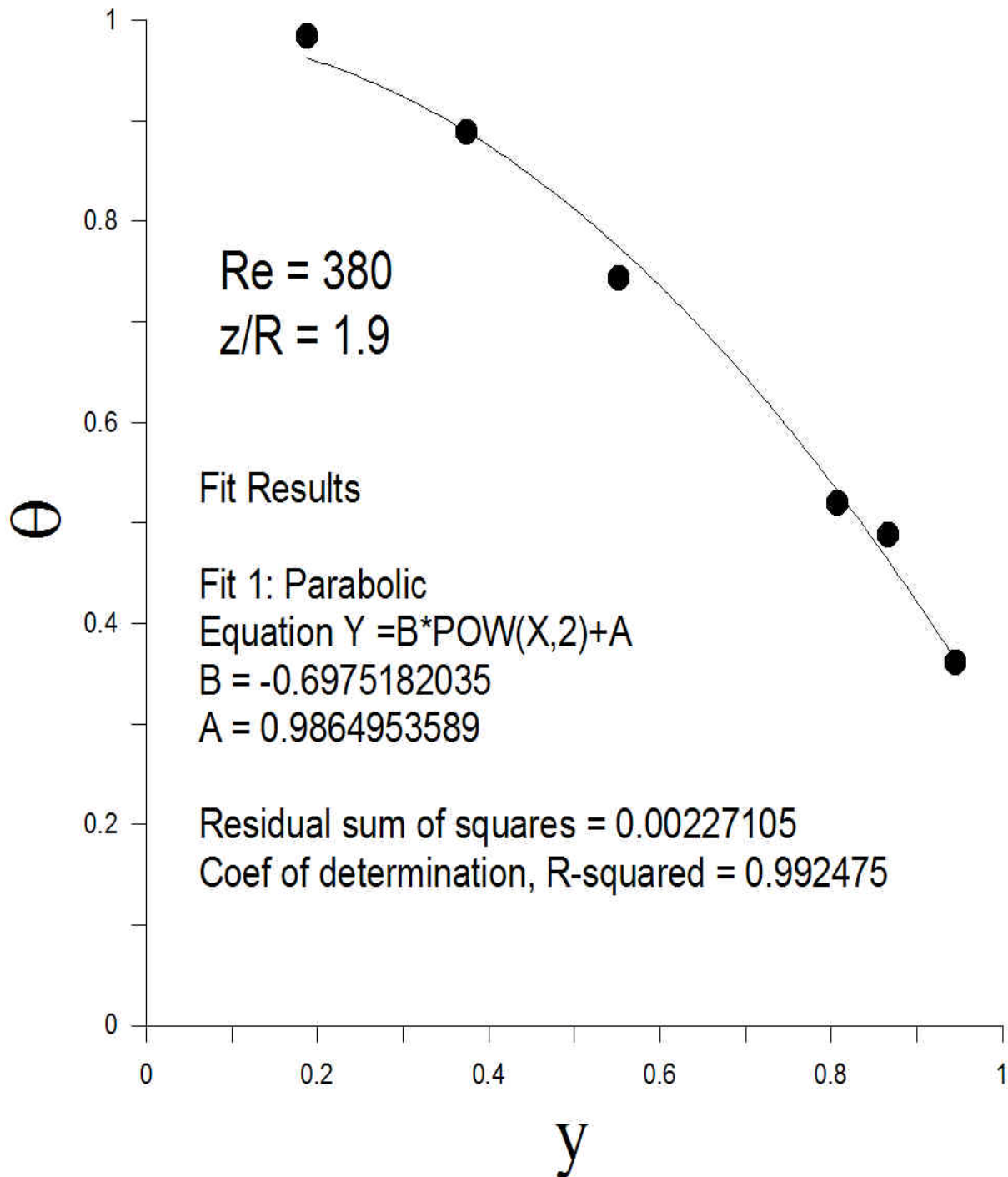
# First Height Parabolic Fit for 4-hole cylinders

In the 4inch Column



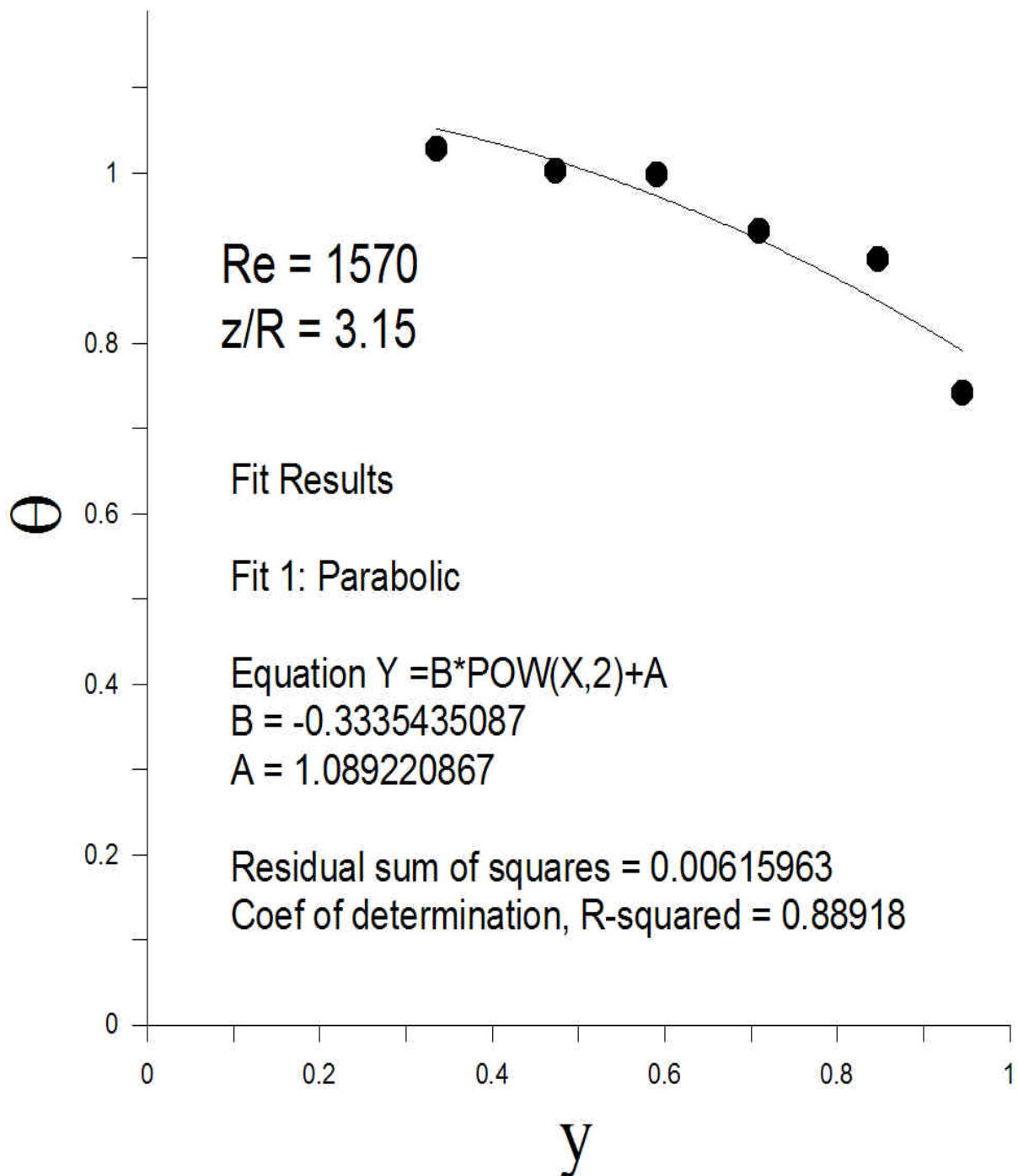
# First Height Parabolic Fit for Monoliths

In the 4inch Column



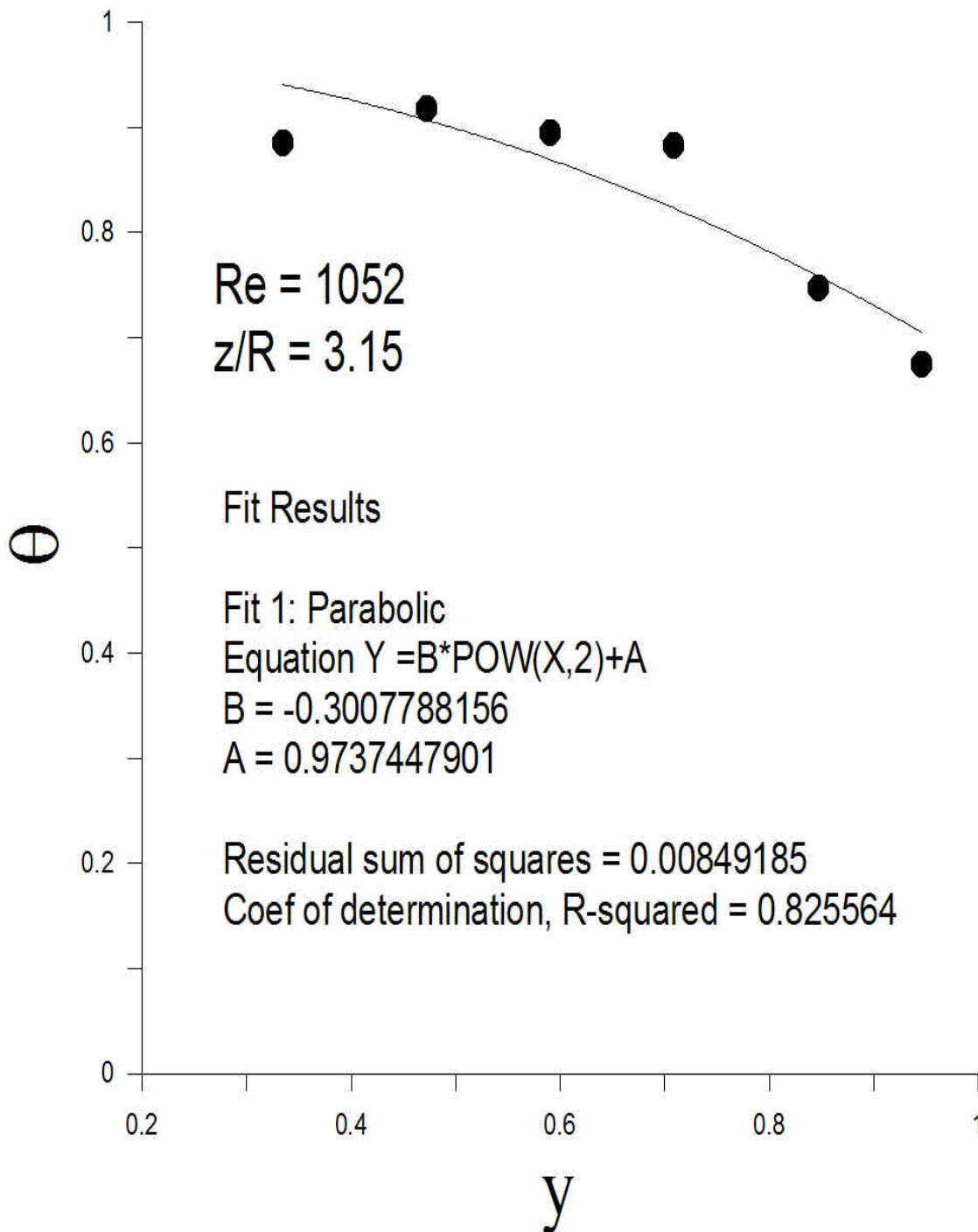
# First Height Parabolic Fit for Raschig Rings

In the 2inch Column



# First Height Parabolic Fit for 4-hole cylinders

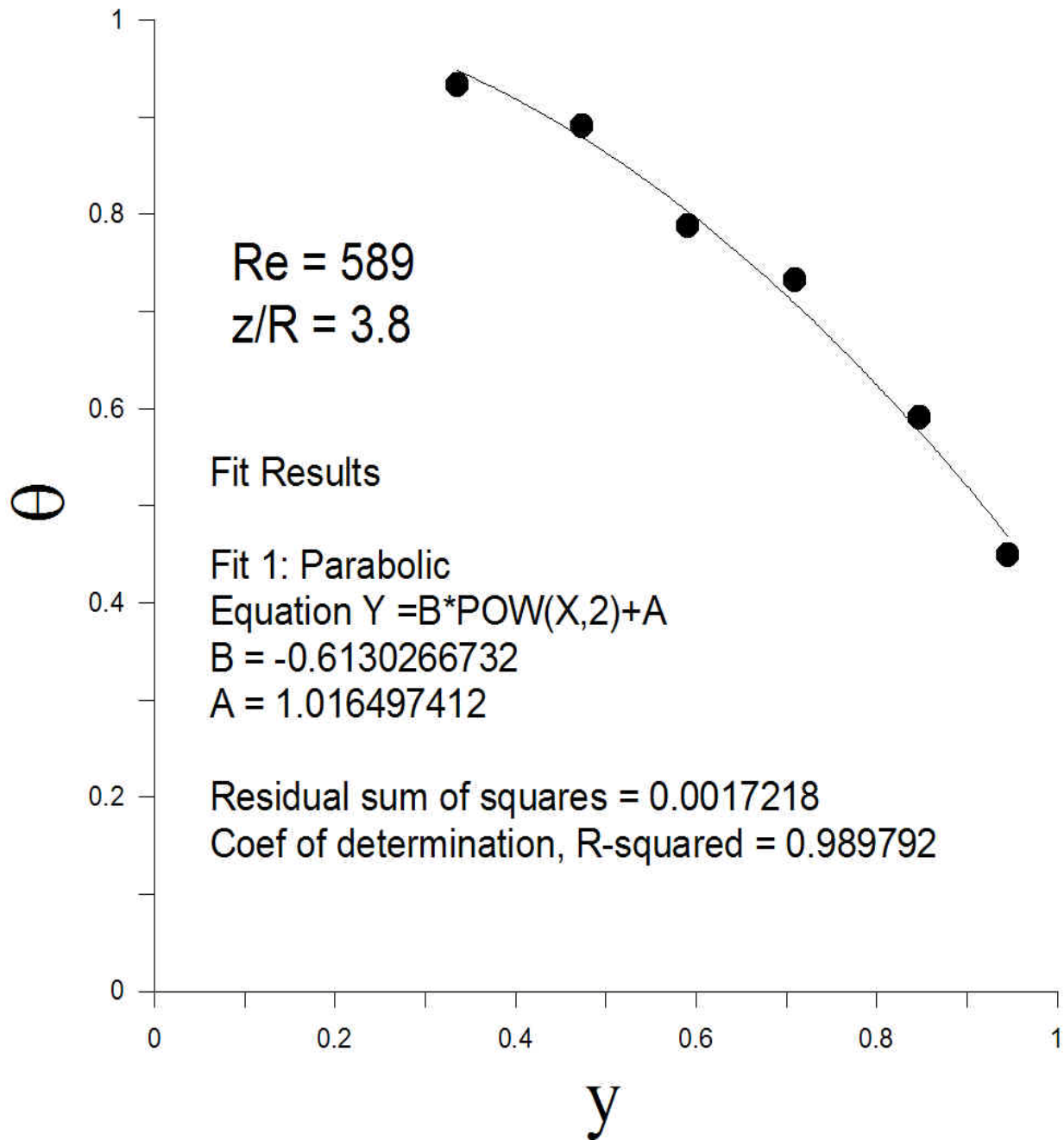
In the 2inch Column





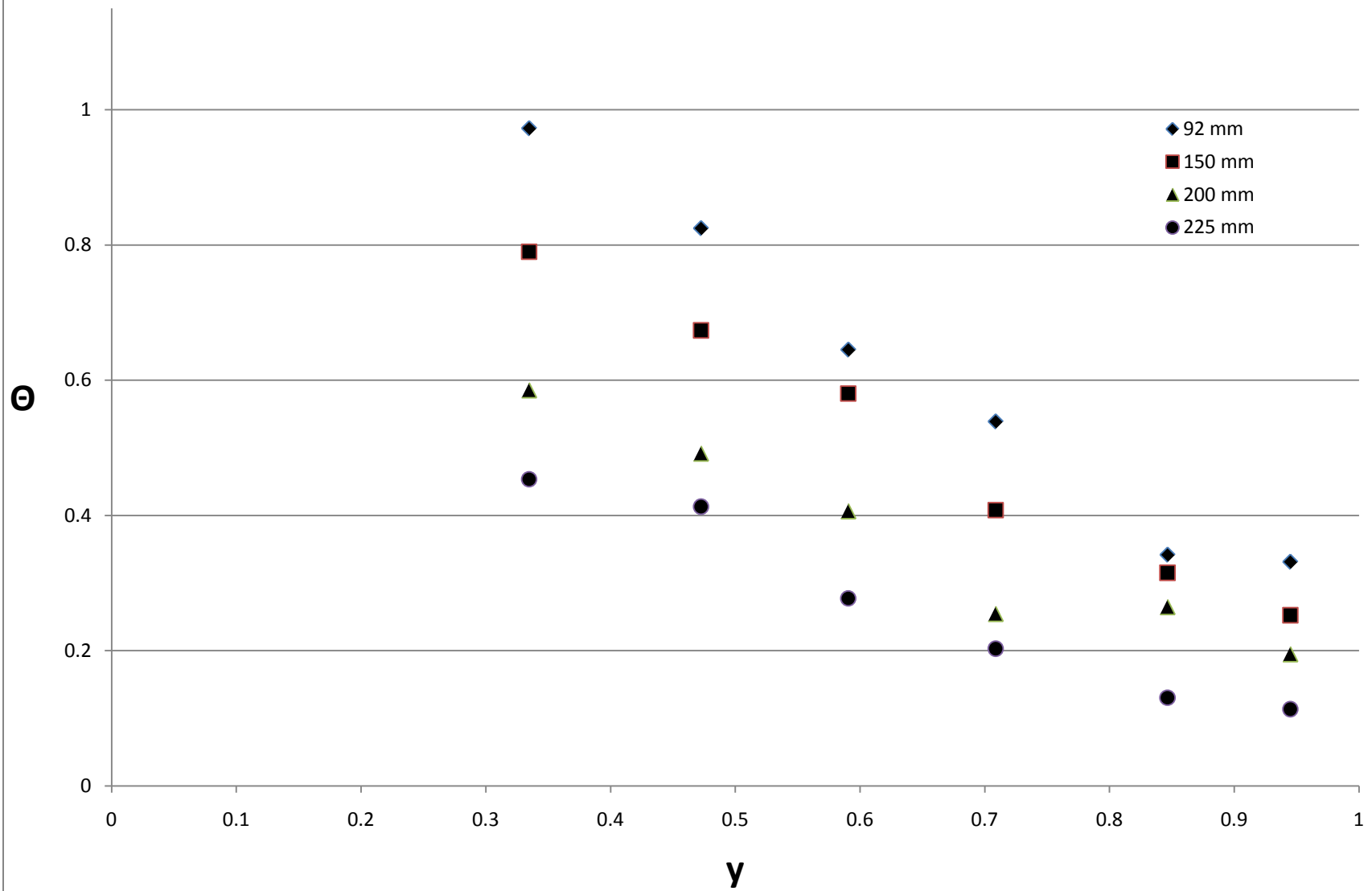
# First Height Parabolic Fit for Monoliths

In the 2inch Column

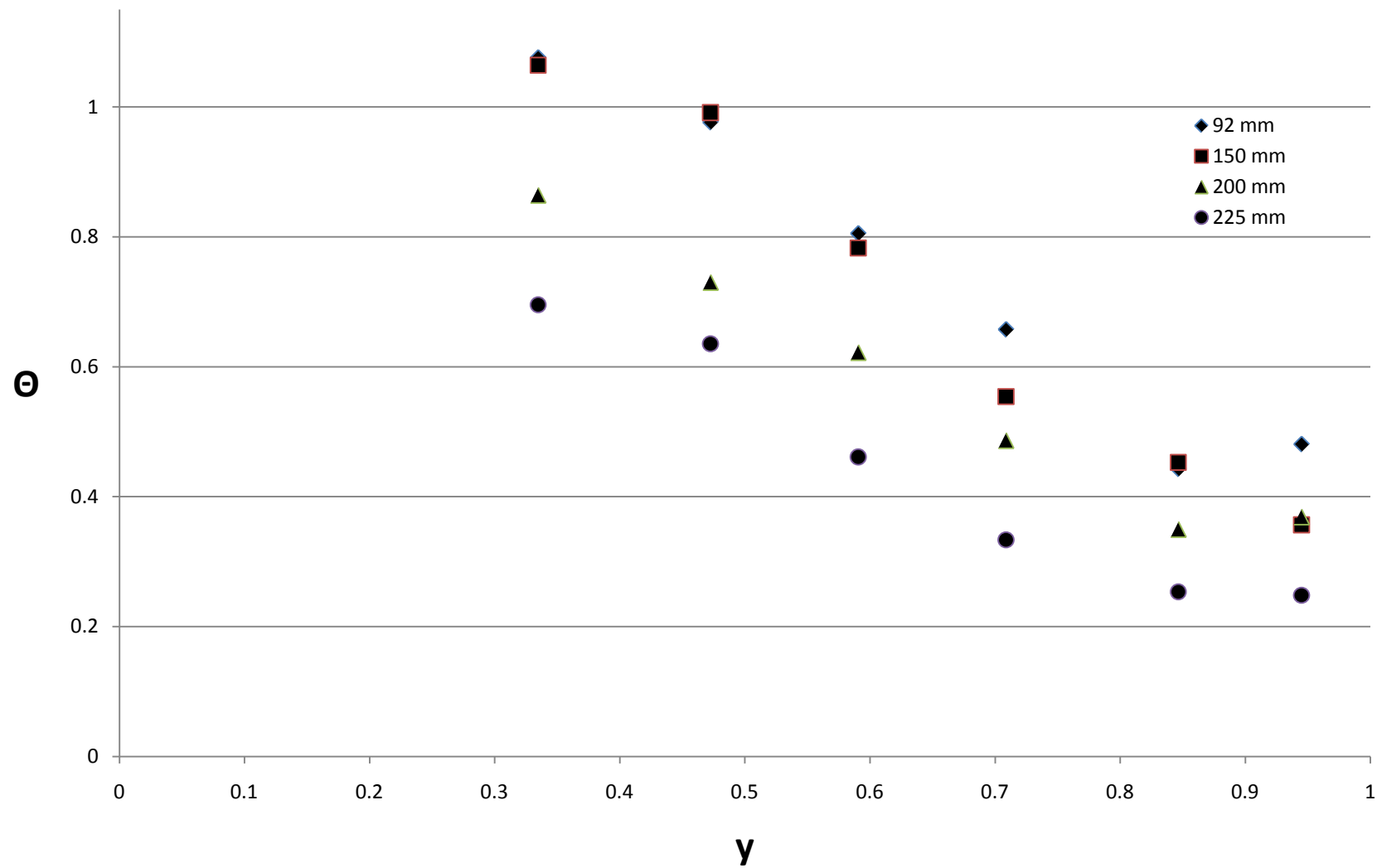


## Appendix C: $\theta$ vs. $y$ by Reynolds Number

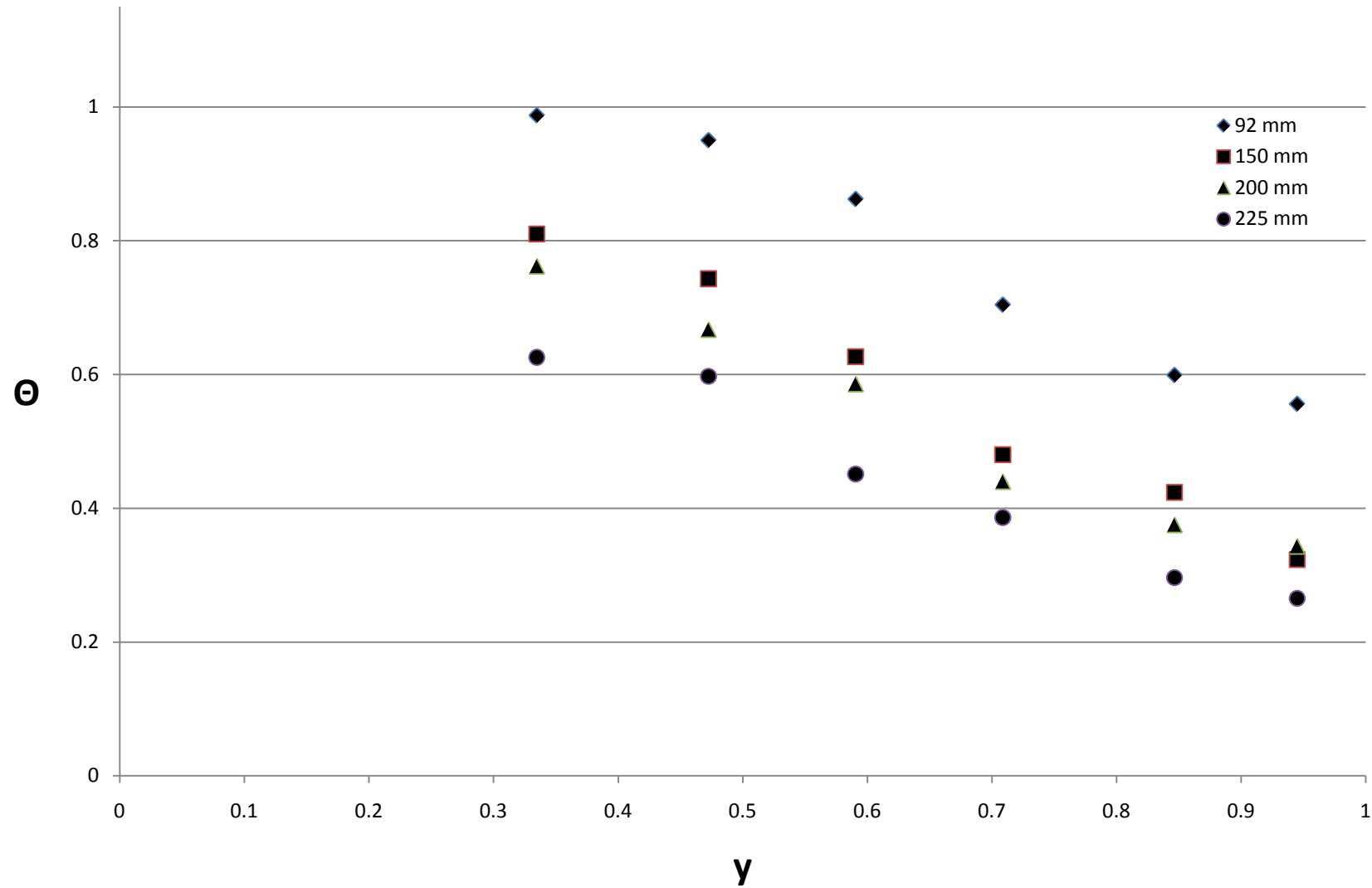
### 4 inch column 0.5 inch spheres Re 75



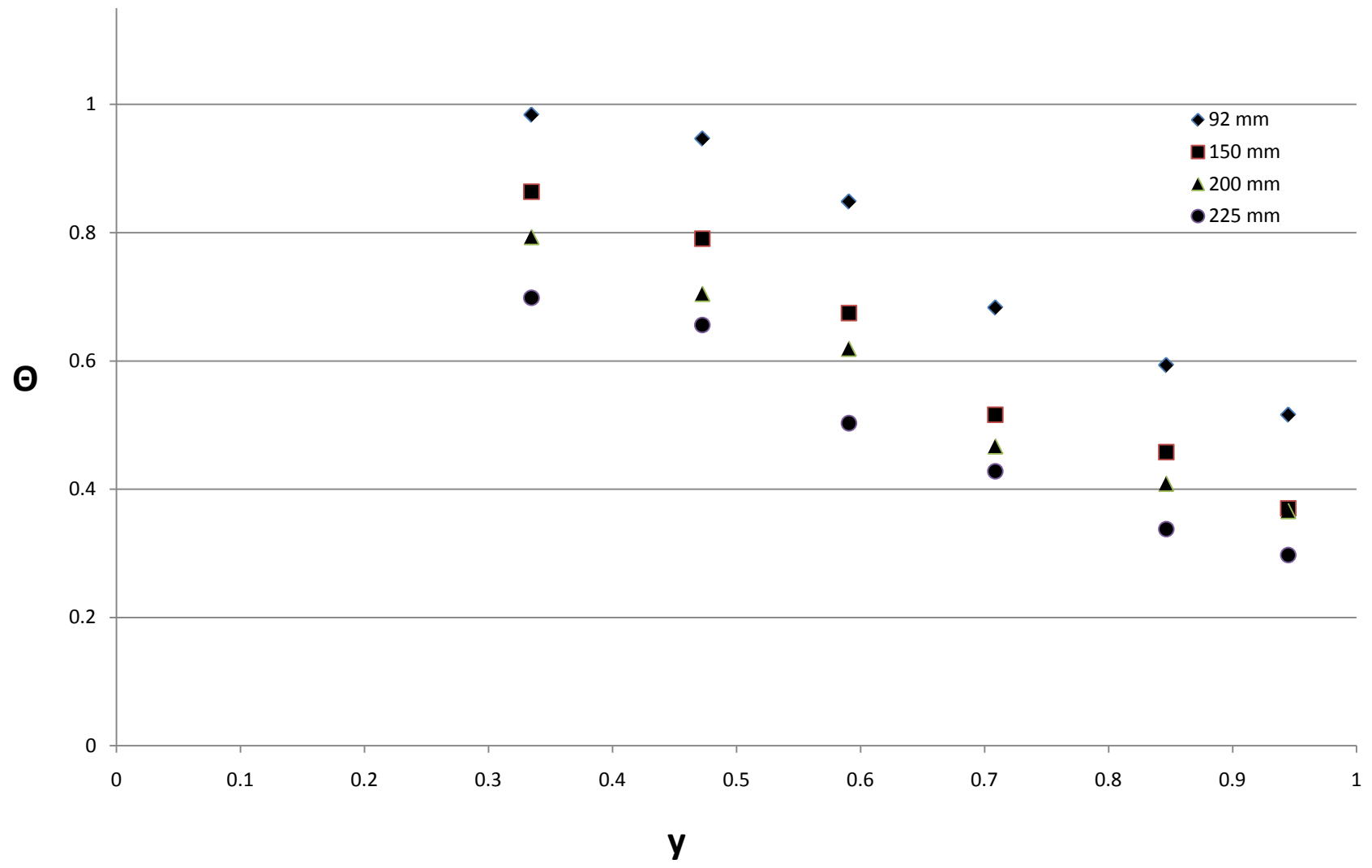
### 4 inch column 0.5 inch spheres Re 145



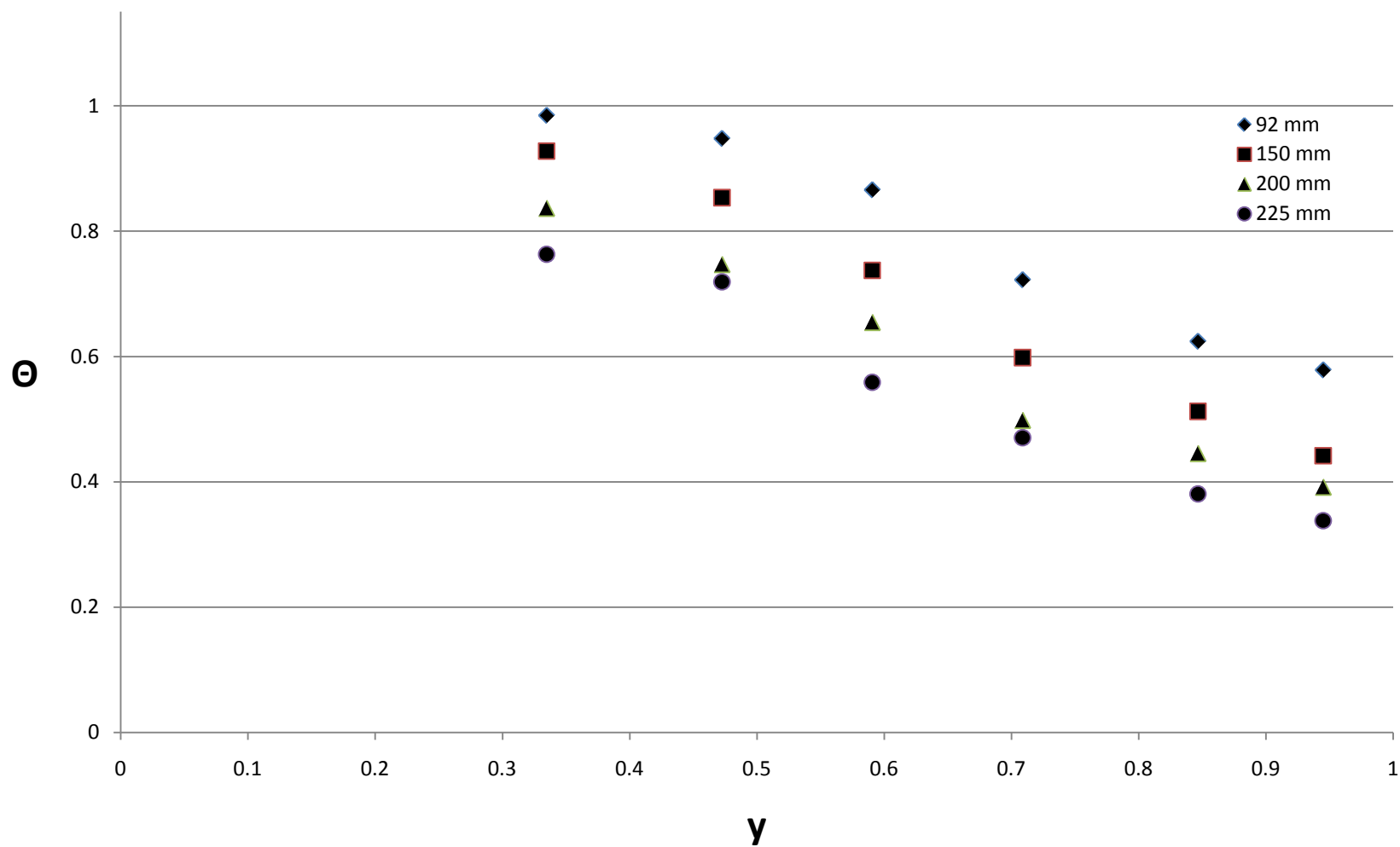
# 4 inch column 0.5 inch spheres Re 265



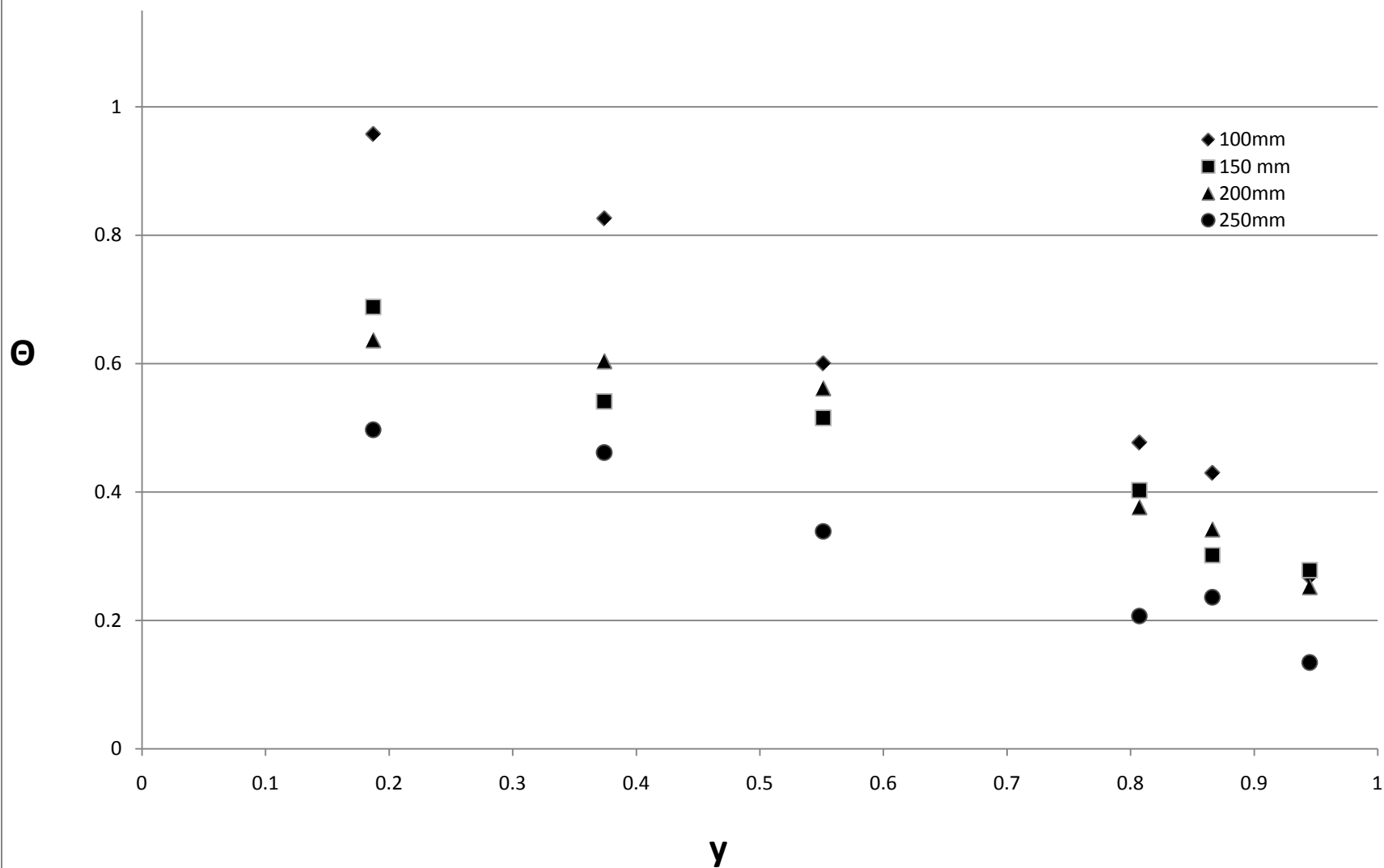
### 4 inch column 0.5 inch spheres Re 335



### 4 inch column 0.5 inch spheres Re 420

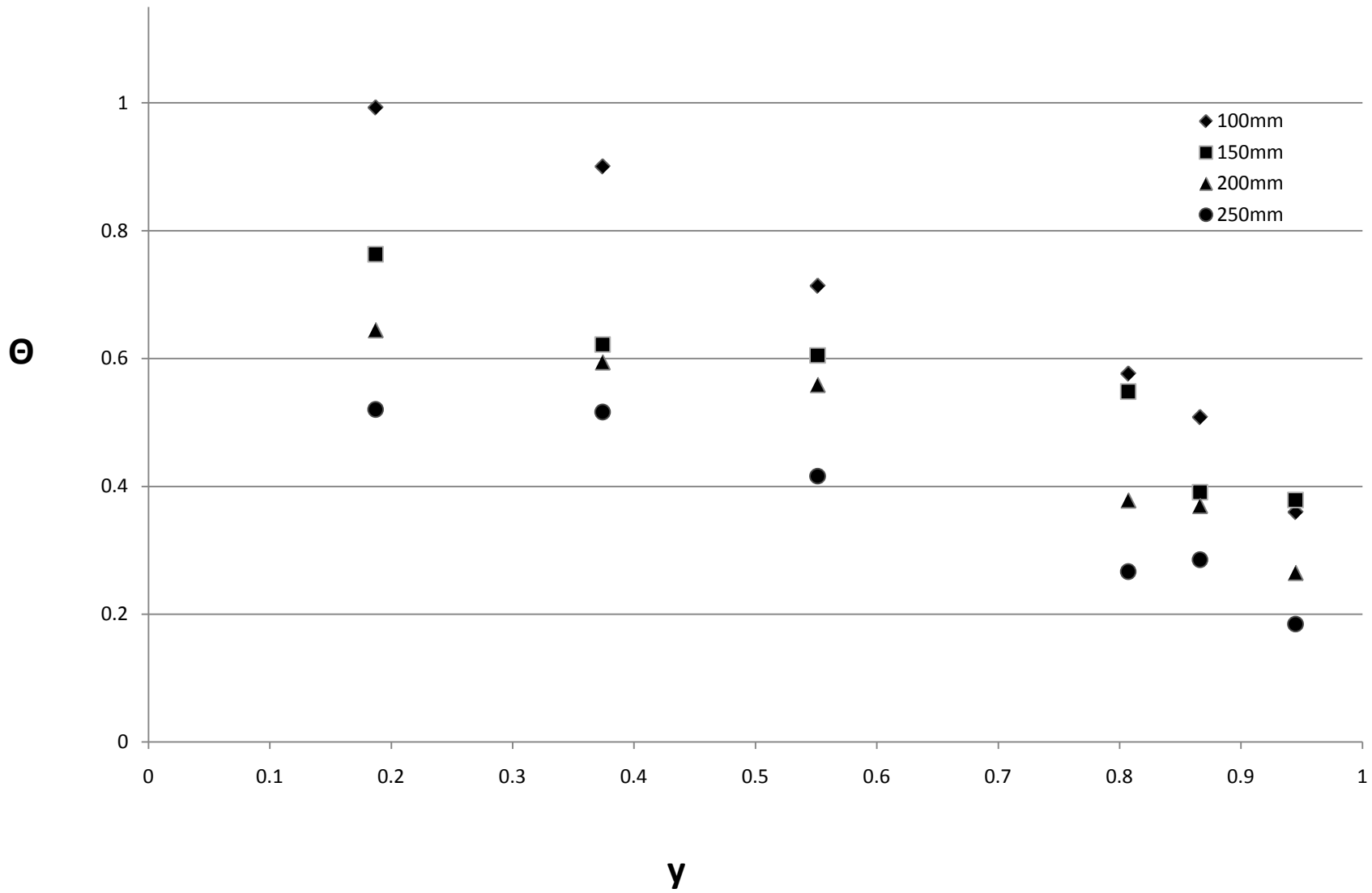


### 4 inch column raschig ring re 85

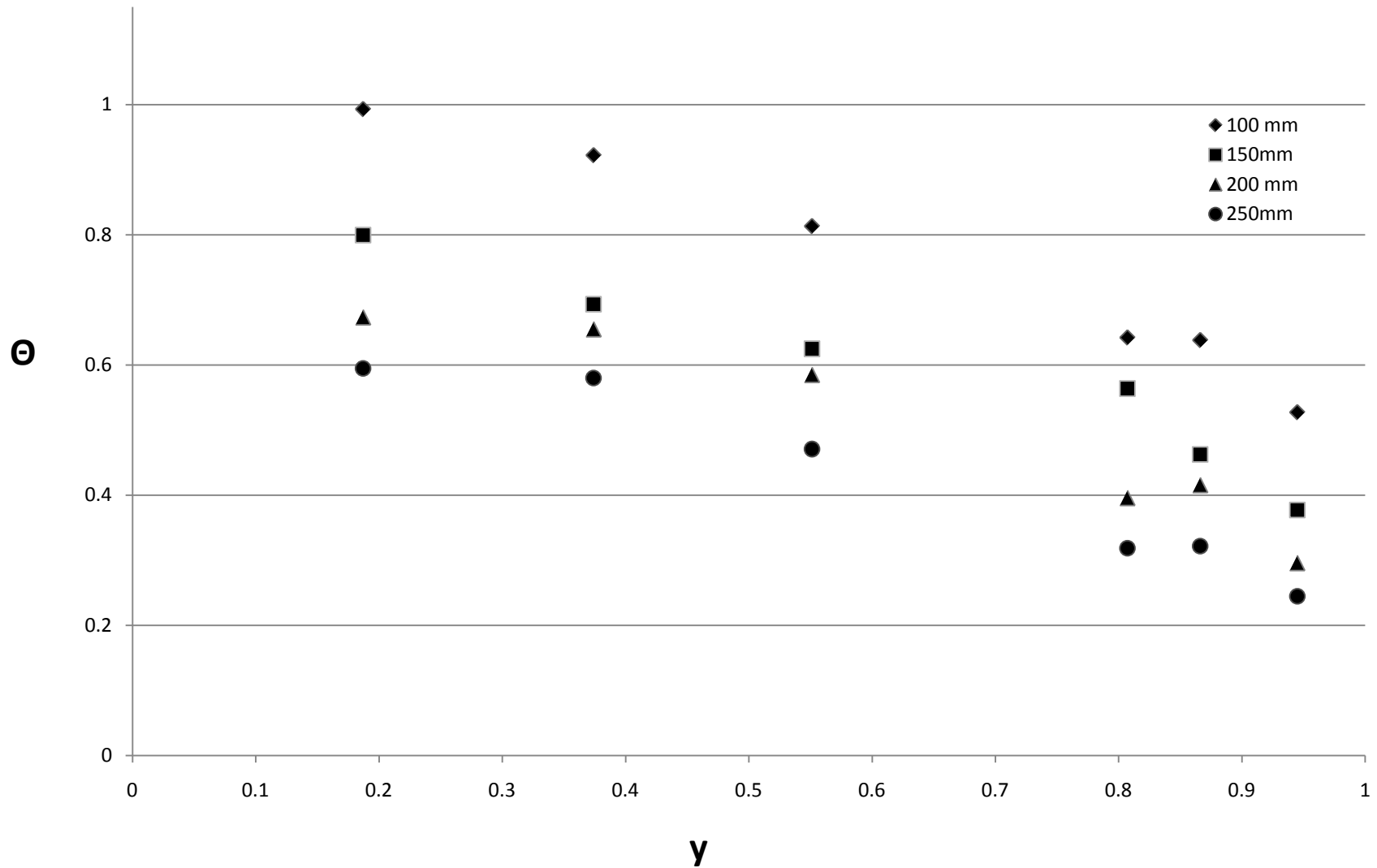




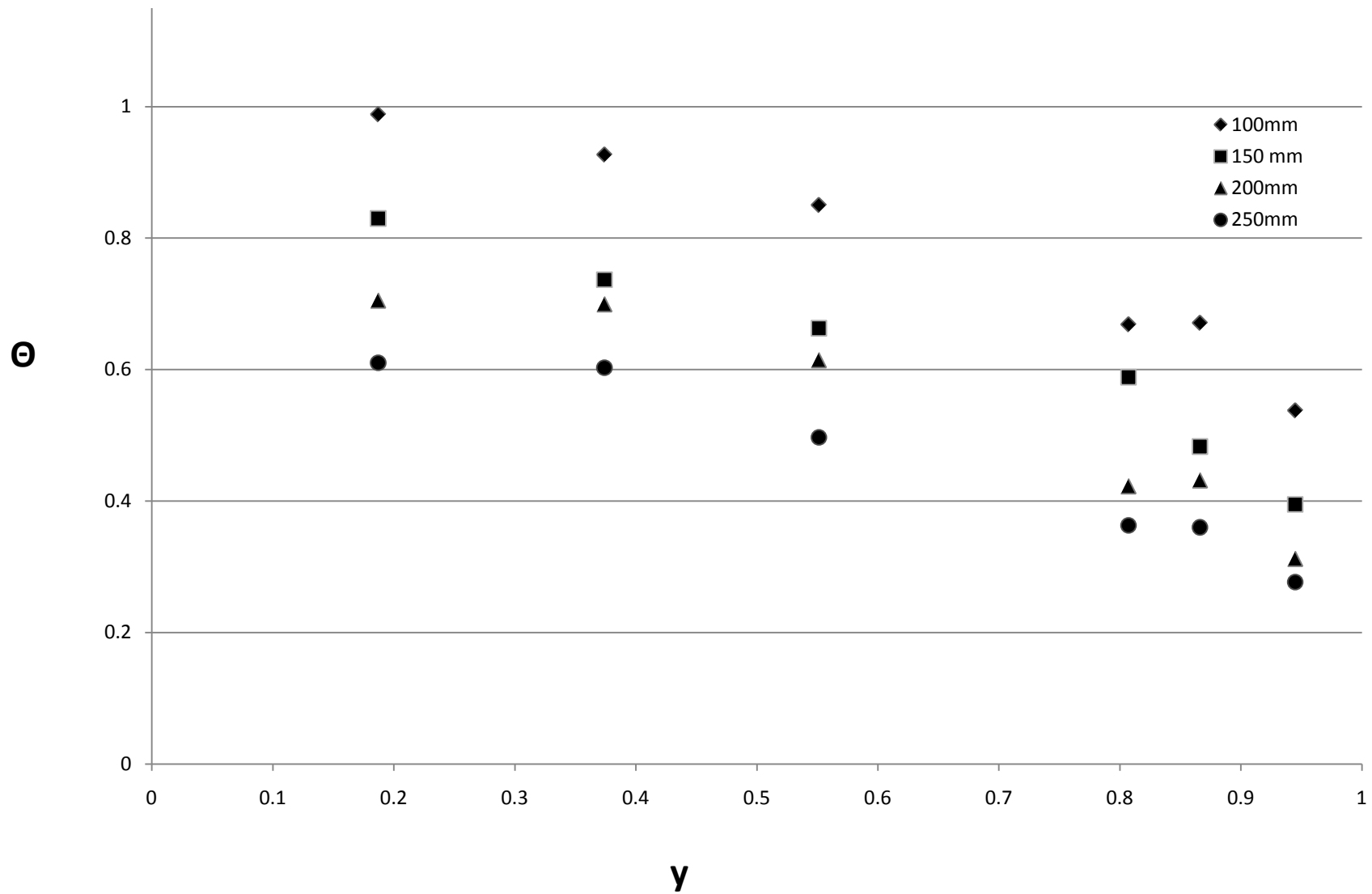
### 4 inch column raschig ring Re 142



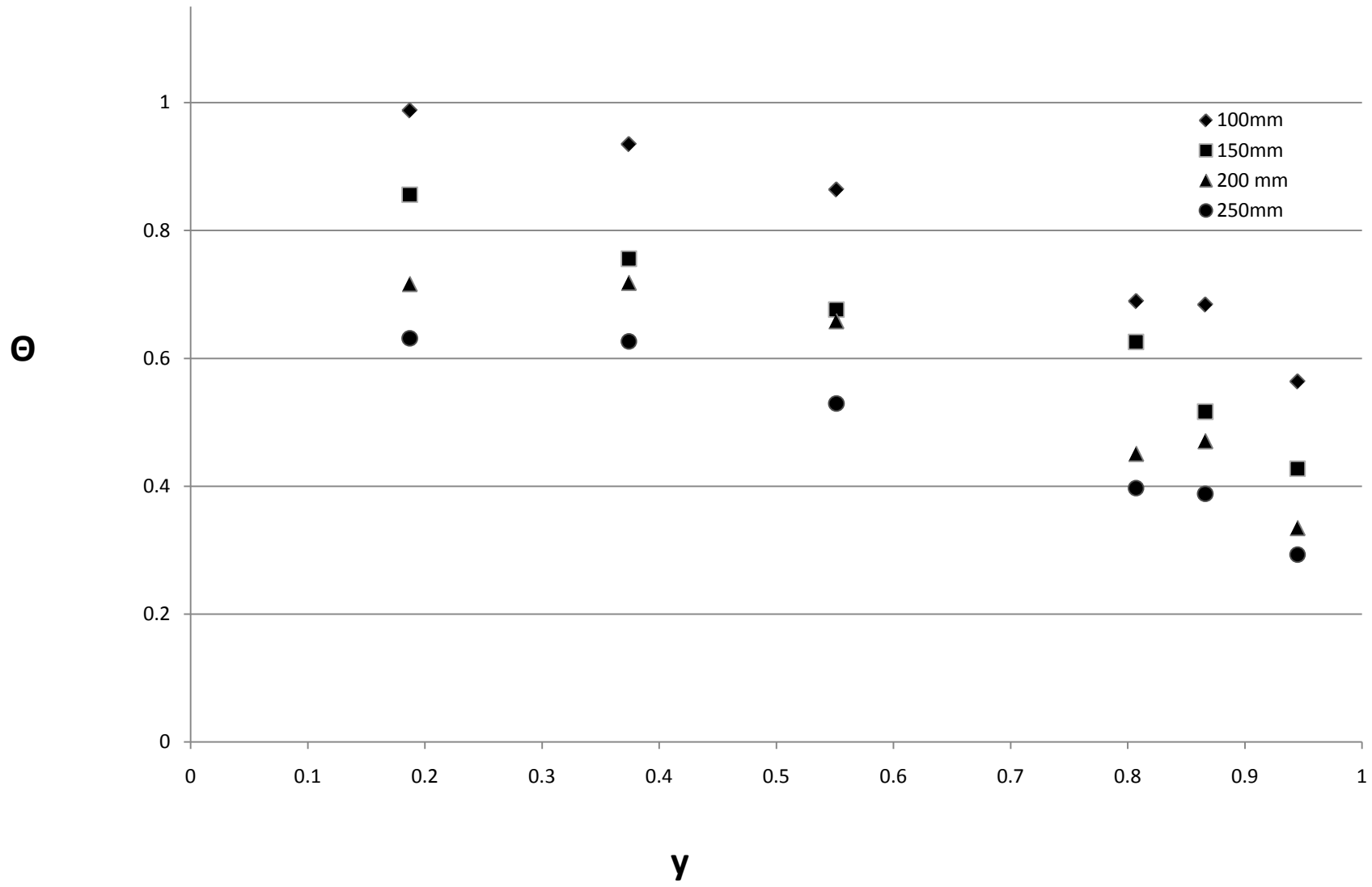
### 4 inch column raschig ring Re 190



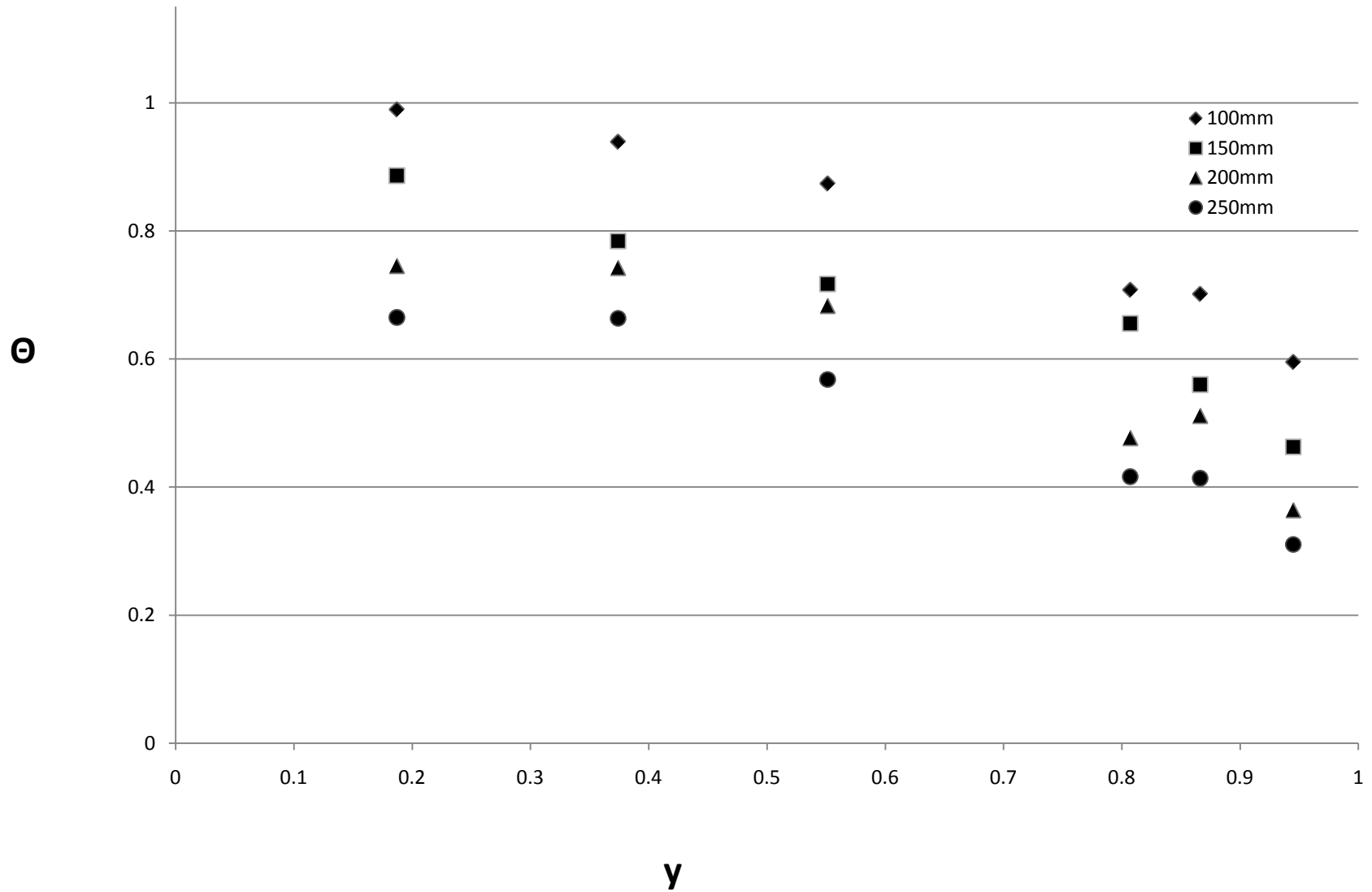
### 4 inch column raschig ring Re 258



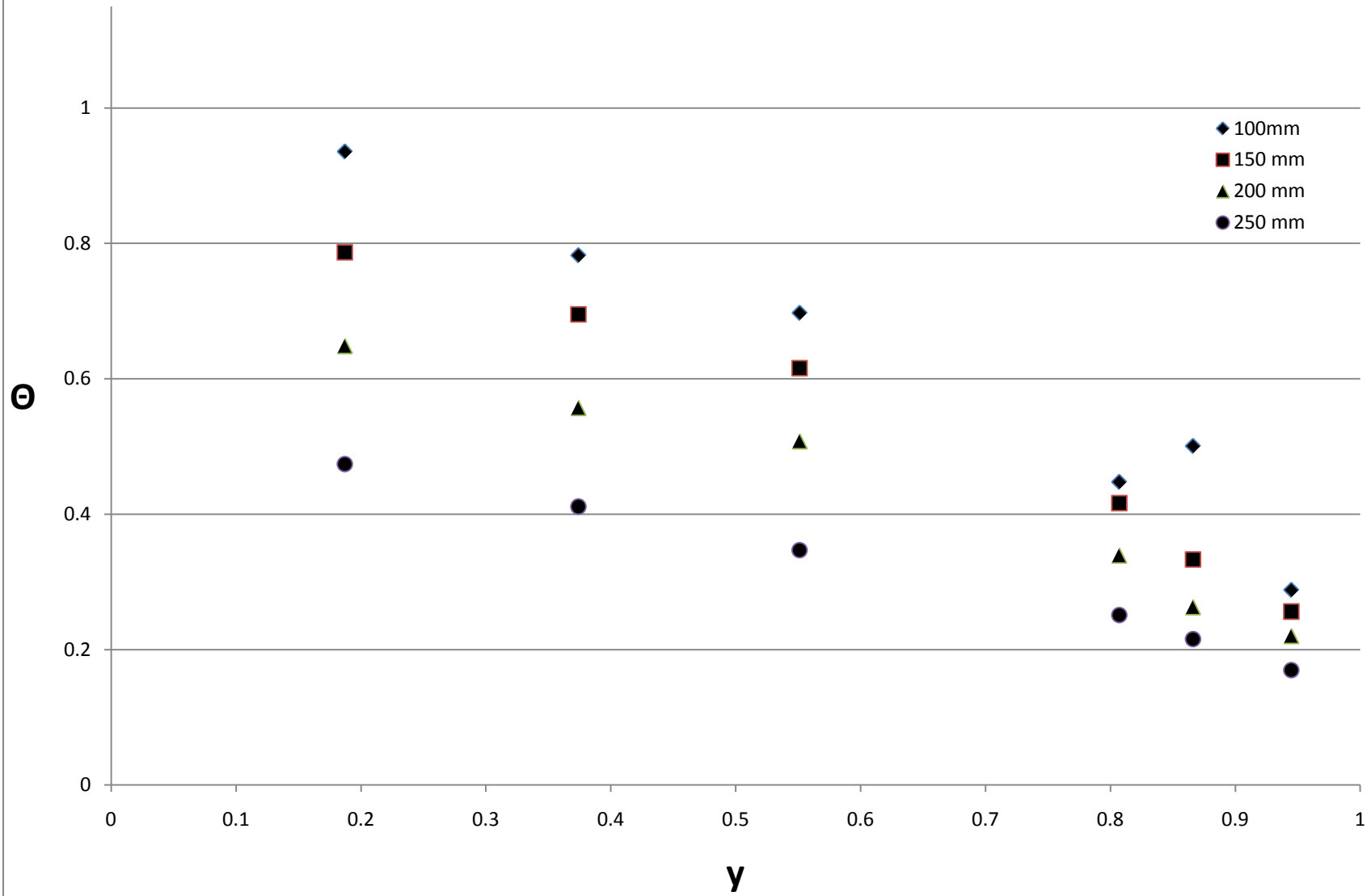
### 4 inch column raschig ring Re 341



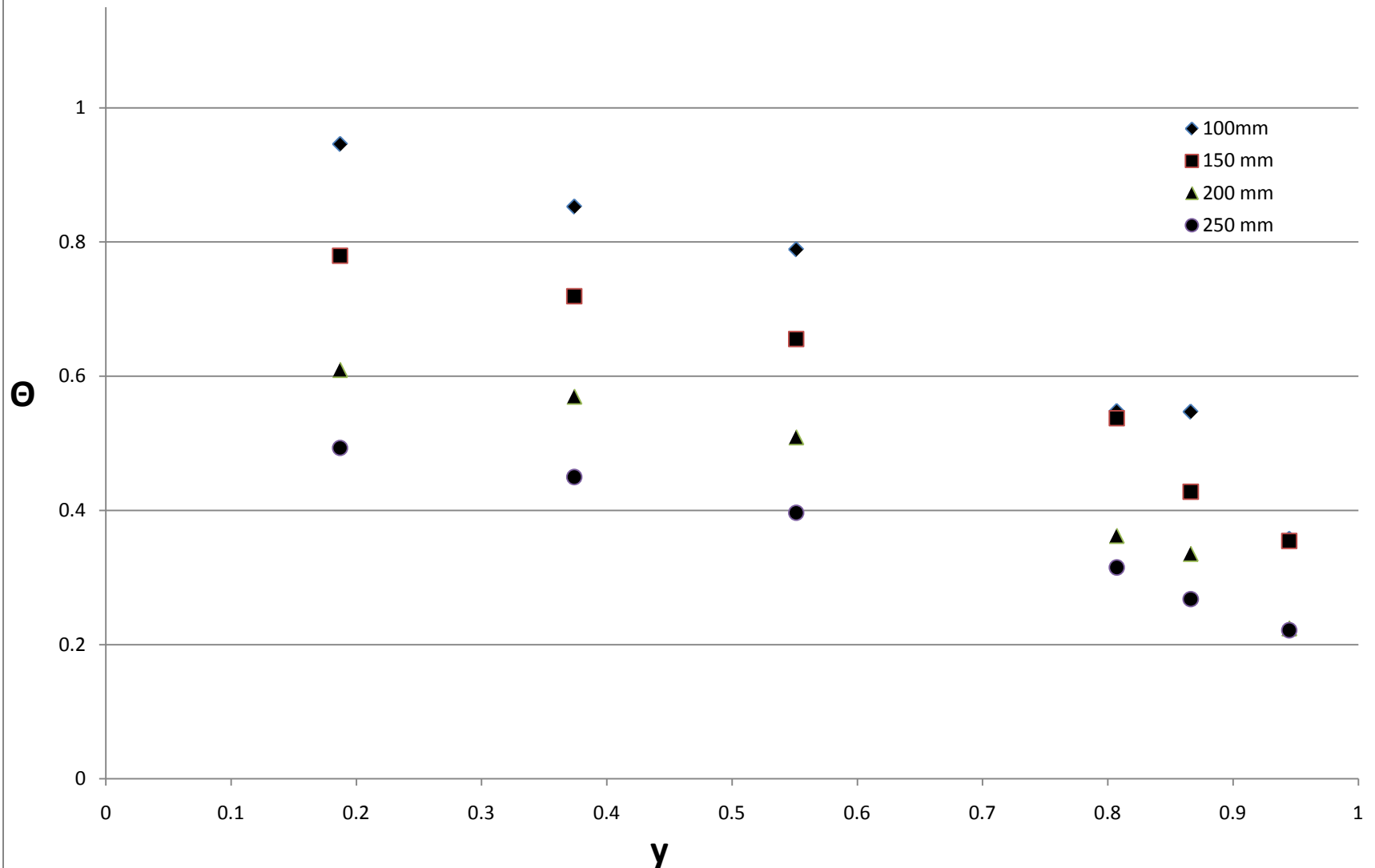
### 4 inch column raschig ring Re 432



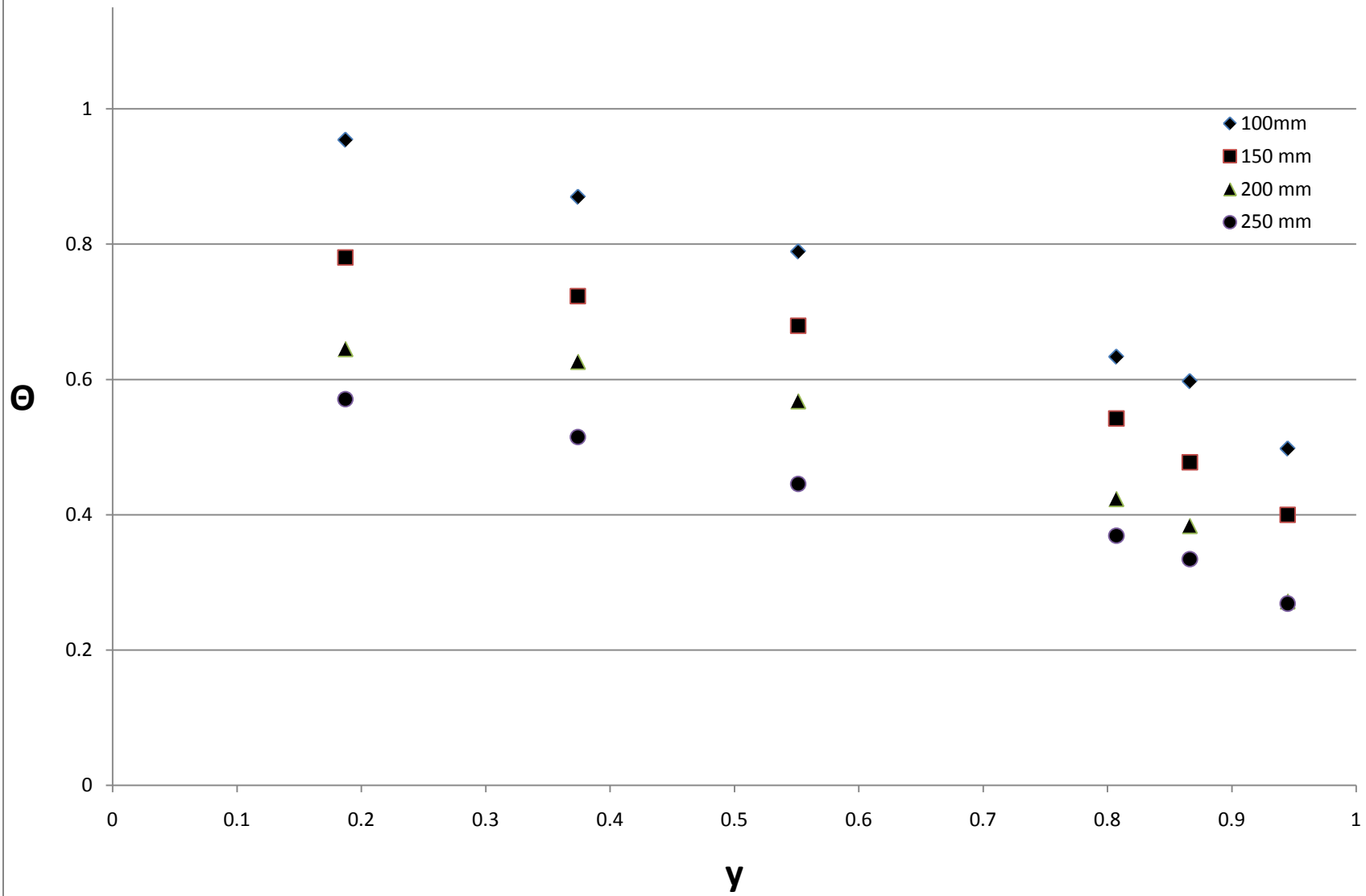
### 4 inch column JM 4-hole cylinders Re 102



### 4 inch column JM 4-hole cylinders Re 198

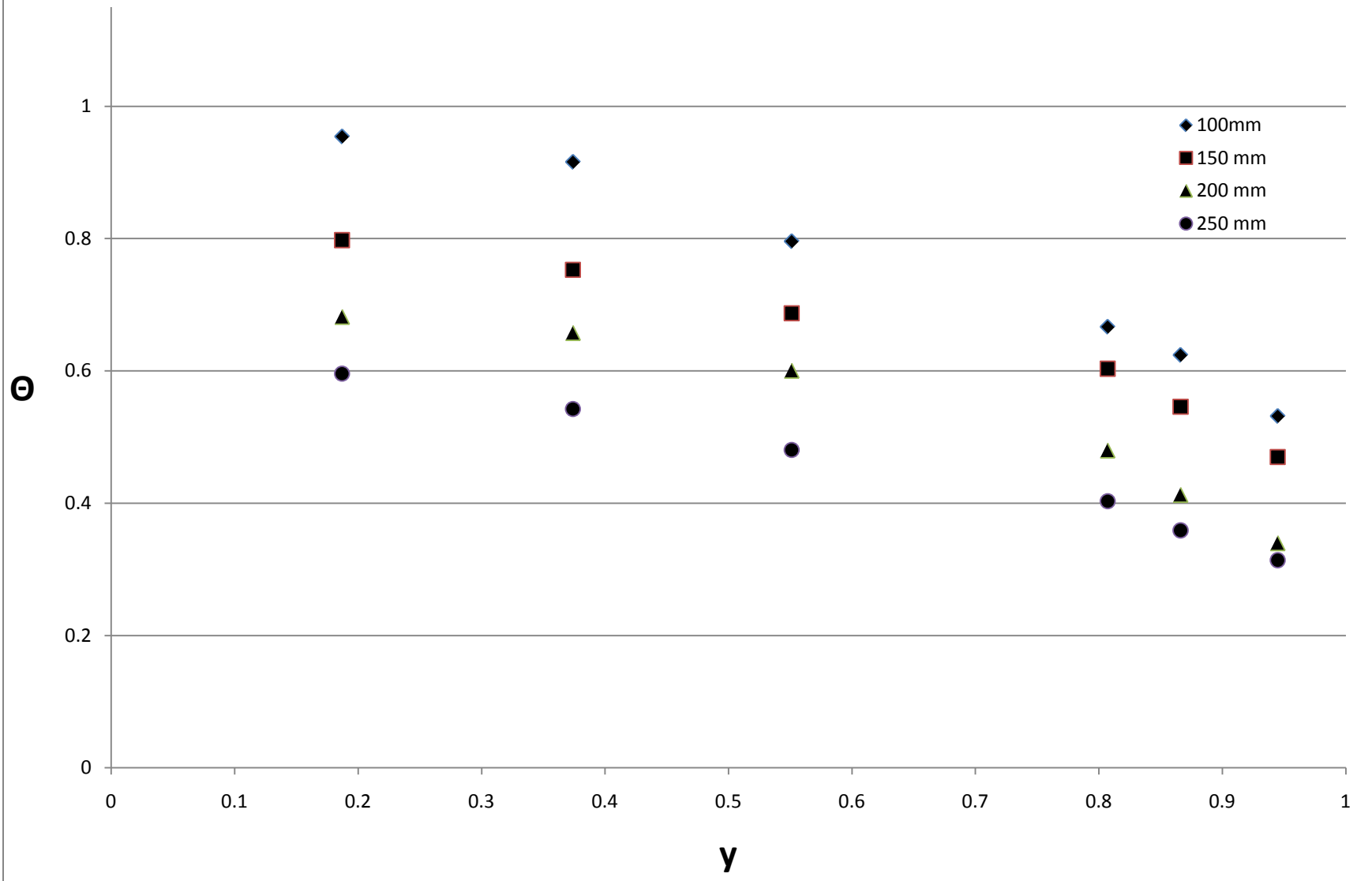


### 4 inch column JM 4-hole cylinders Re 268

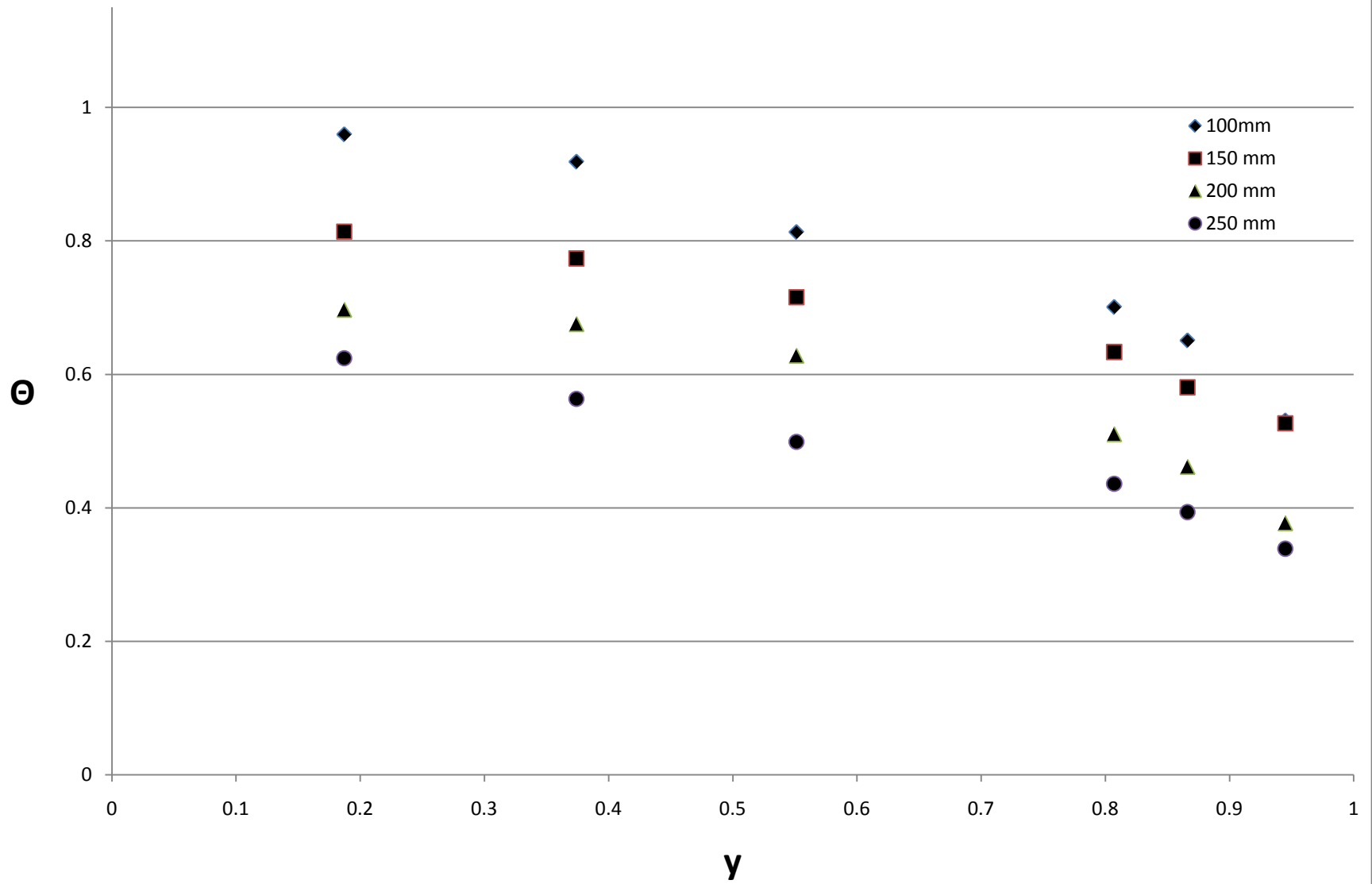




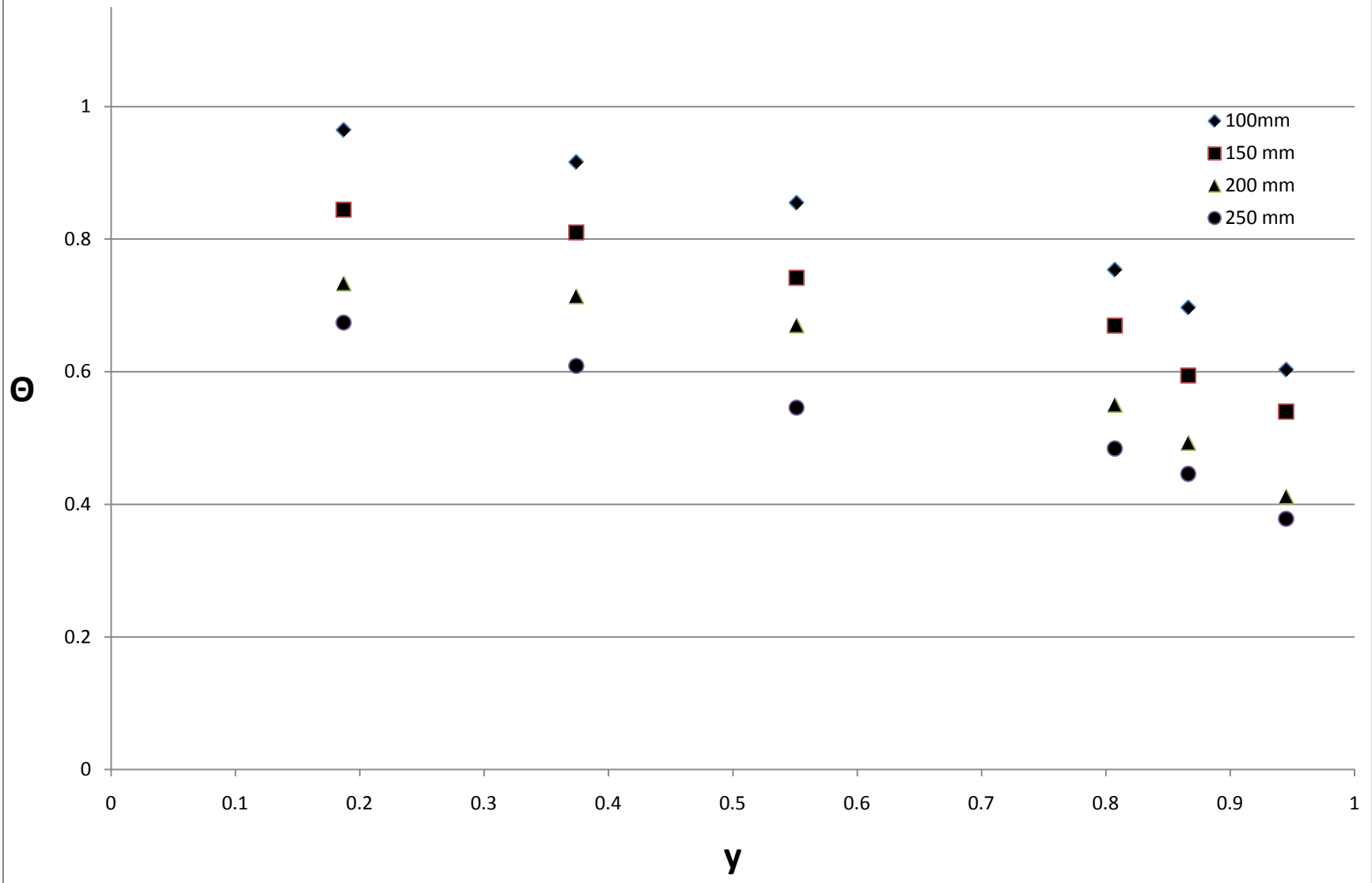
### 4 inch column JM 4-hole cylinders Re 358



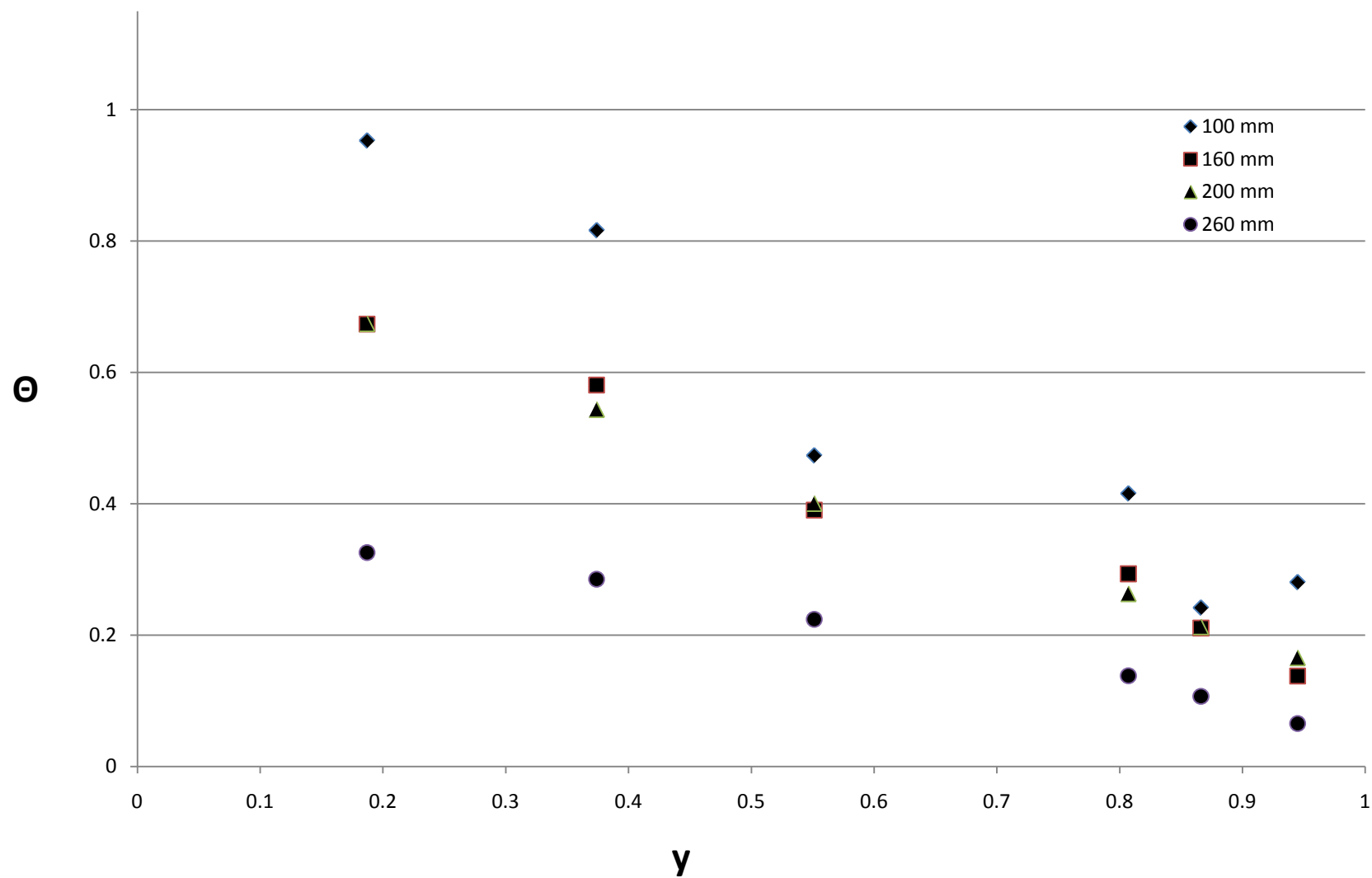
### 4 inch column JM 4-hole cylinders Re 460



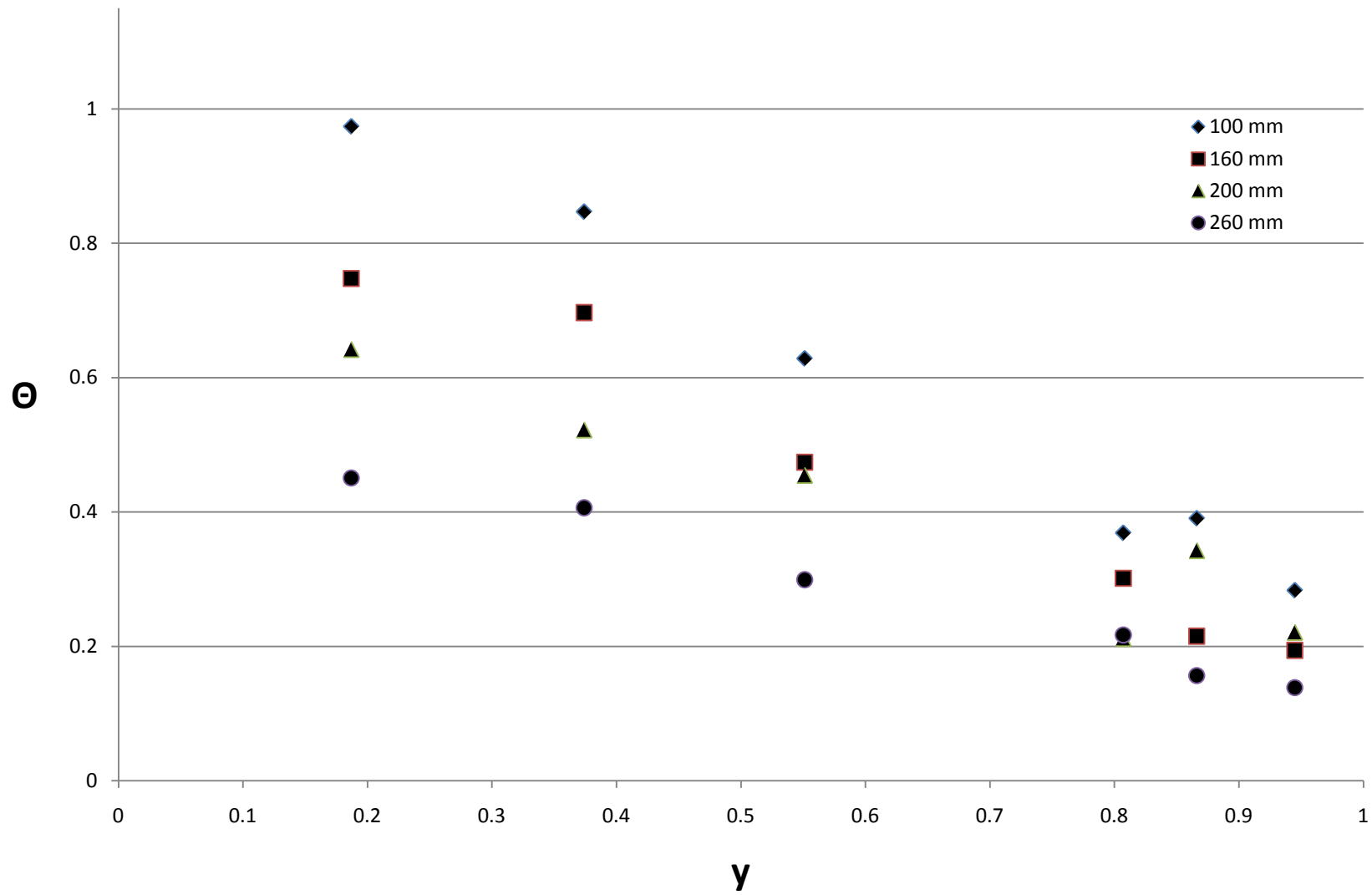
### 4 inch column JM 4-hole cylinders Re 584



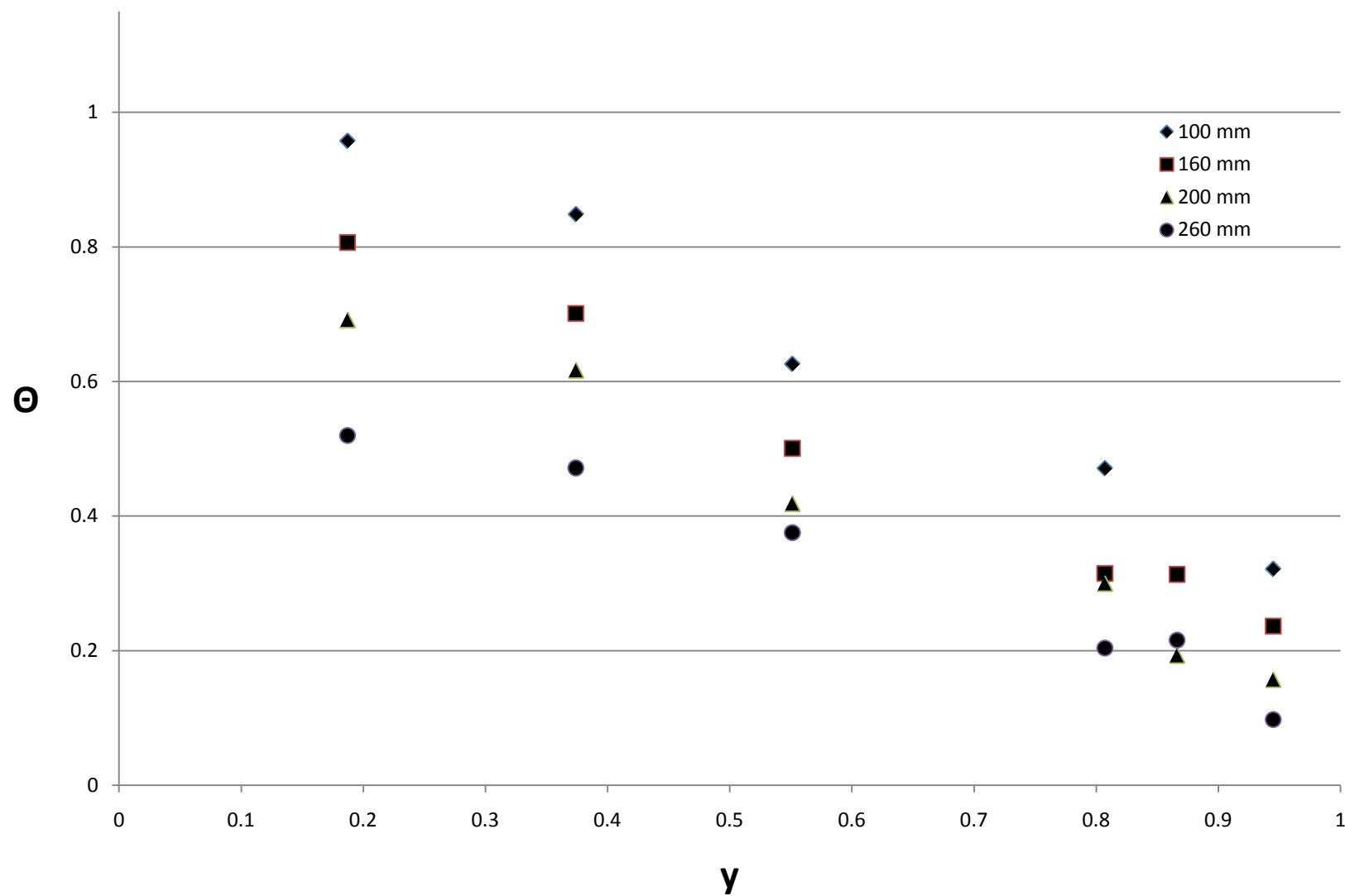
### 4 inch column monoliths Re 43



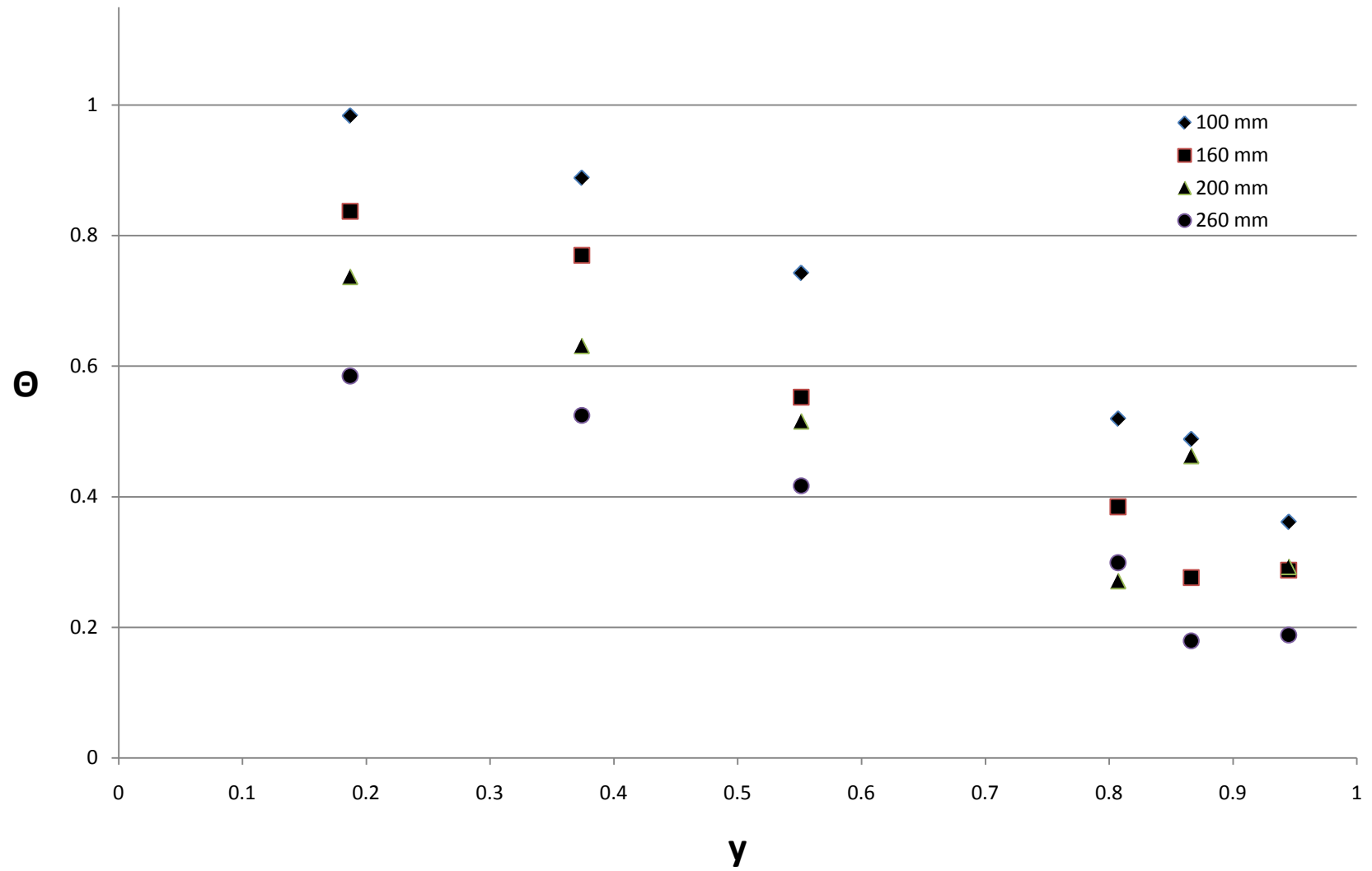
### 4 inch column monoliths Re 71



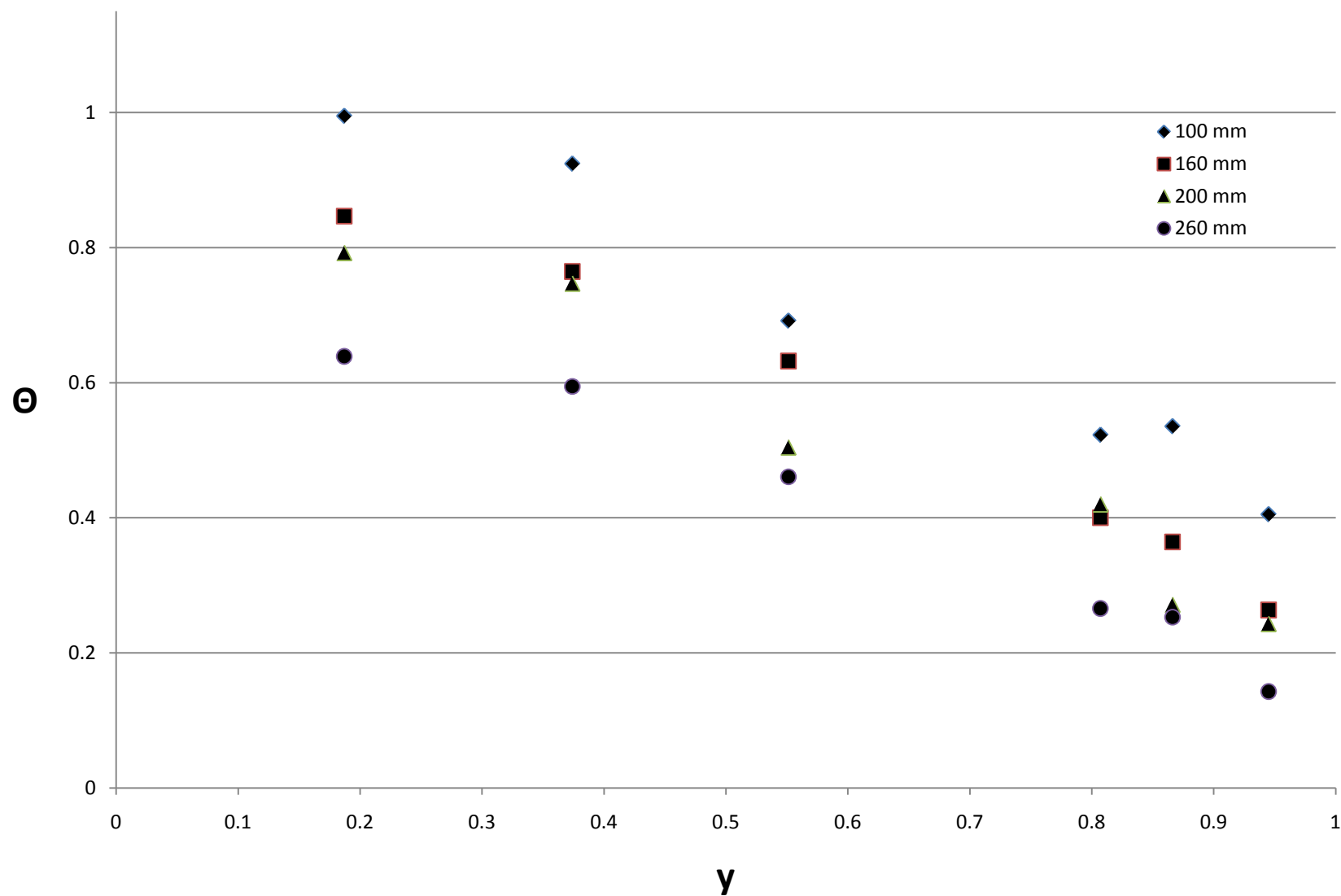
### 4 inch column monoliths Re 95



### 4 inch column monoliths Re 129

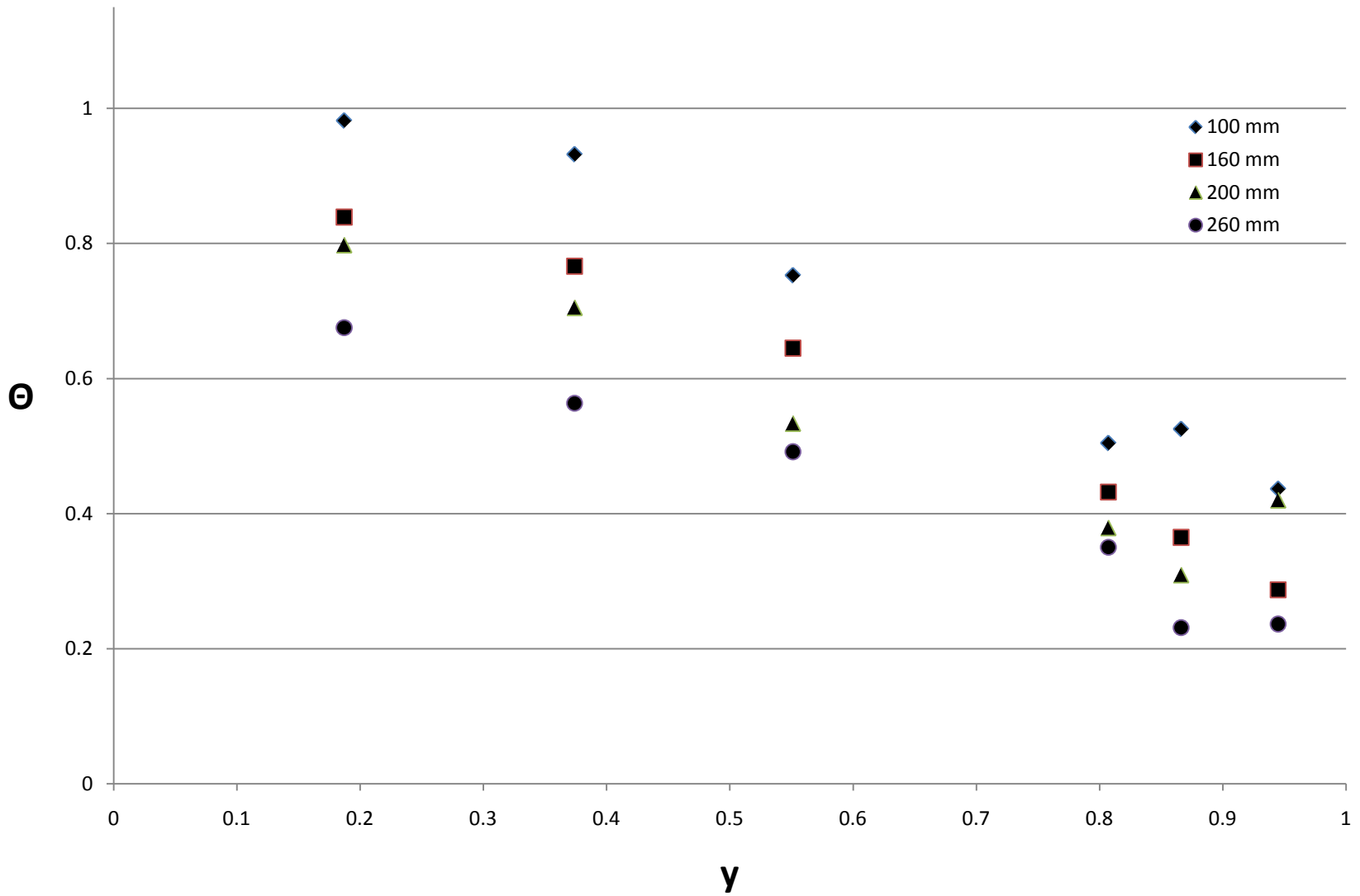


### 4 inch column monoliths Re 171

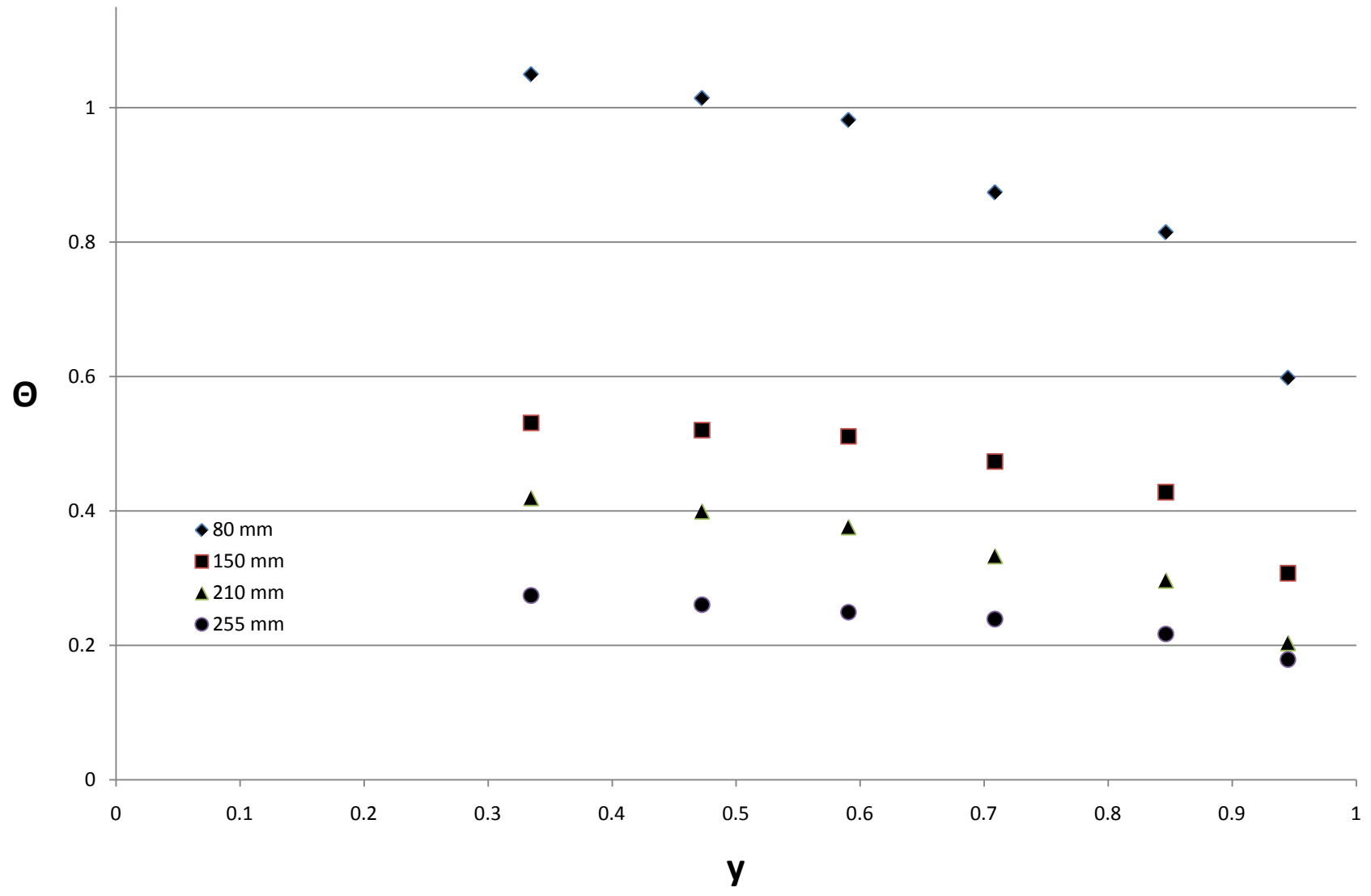




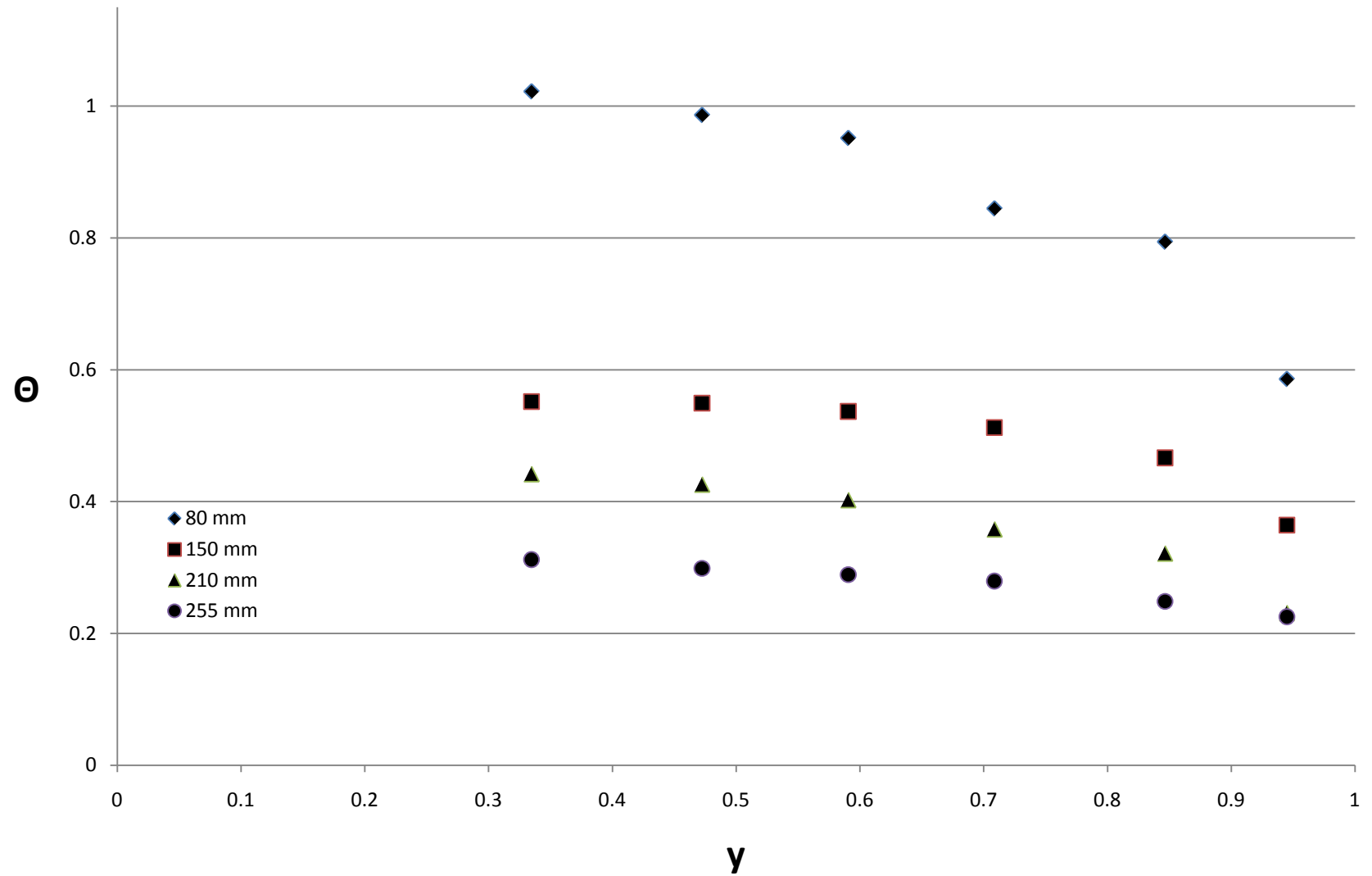
### 4 inch column monoliths Re 216



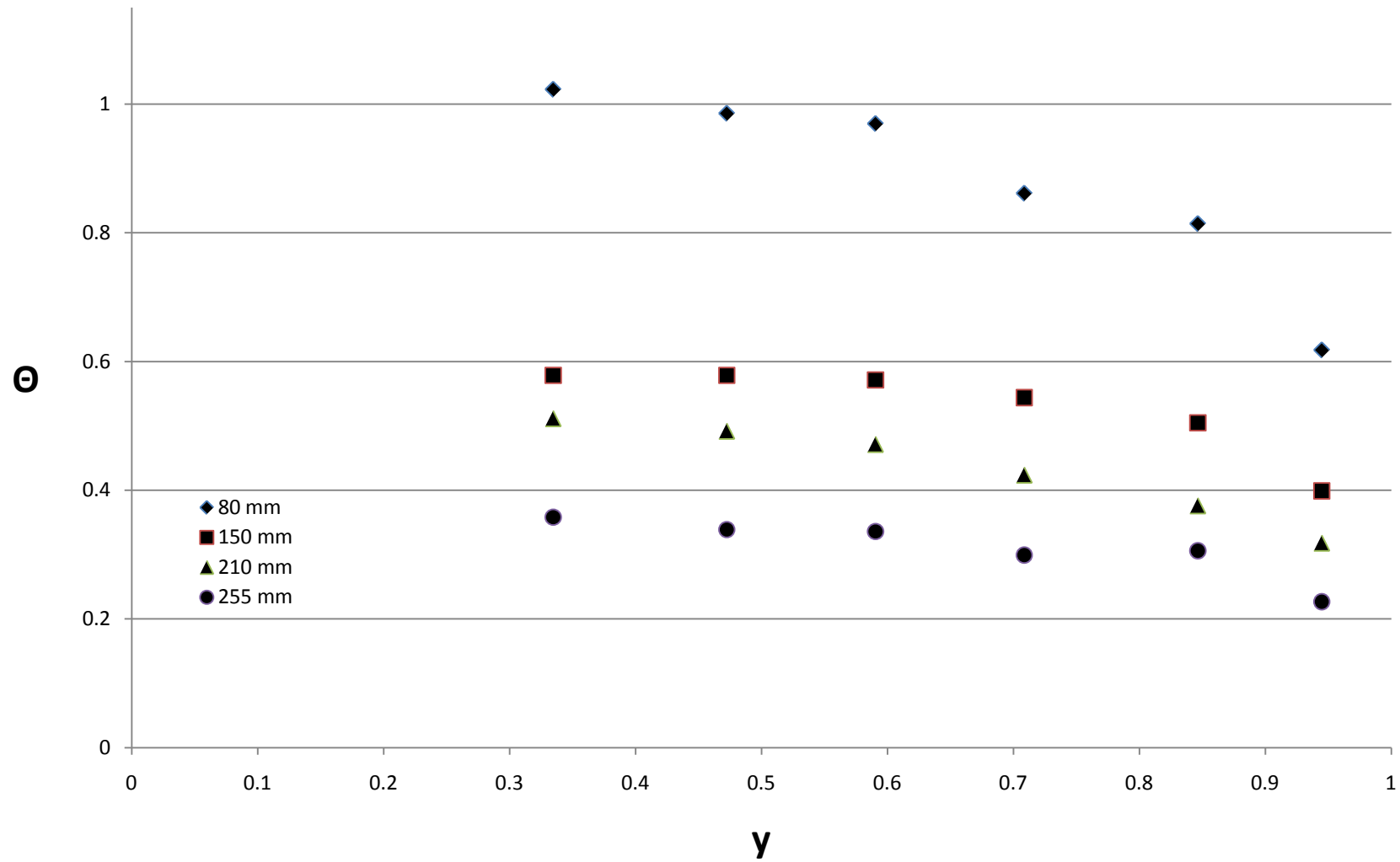
## 2 inch column raschig ring Re 350



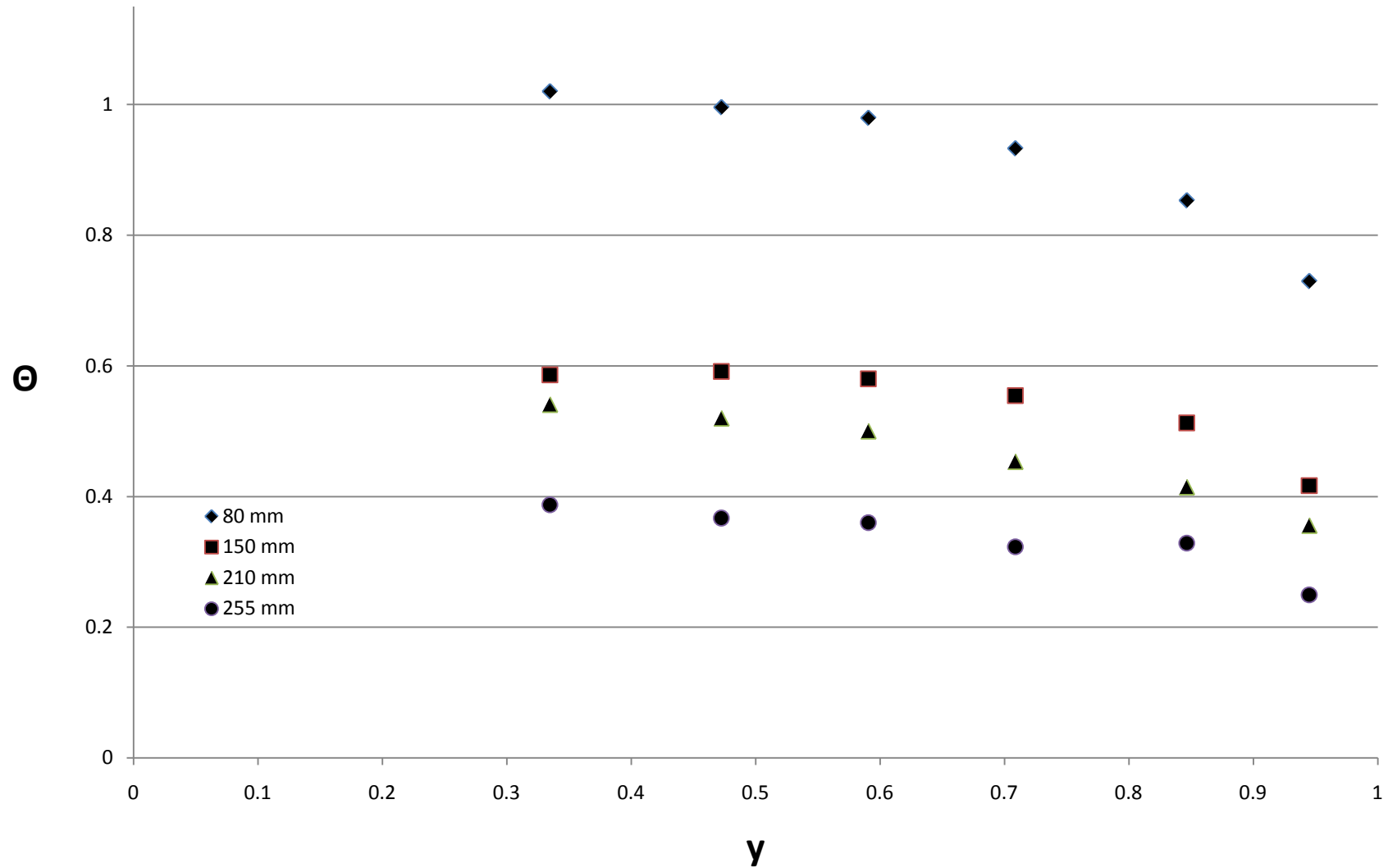
## 2 inch column raschig ring Re 490



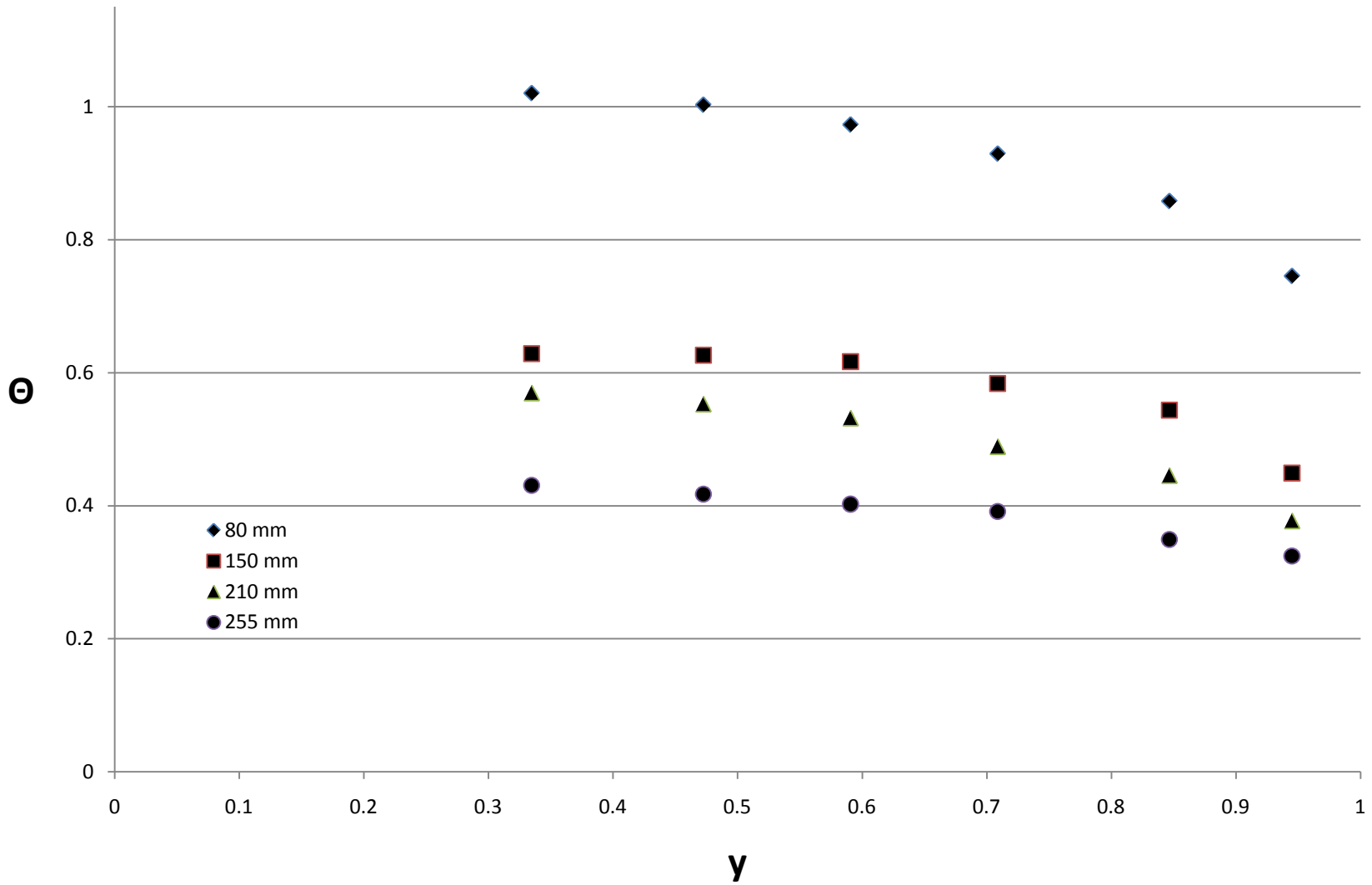
## 2 inch column raschig ring Re 676



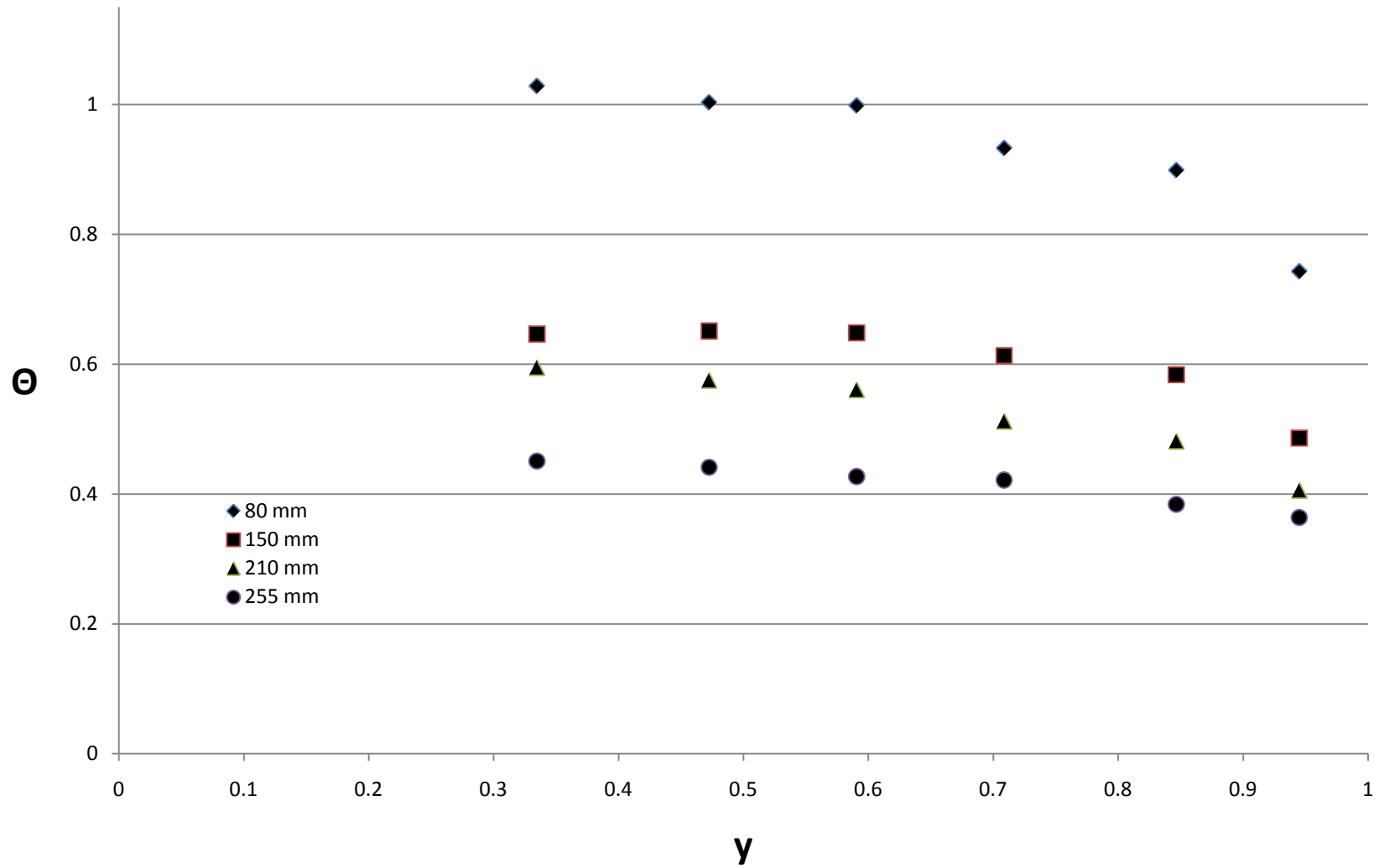
## 2 inch column raschig ring Re 909



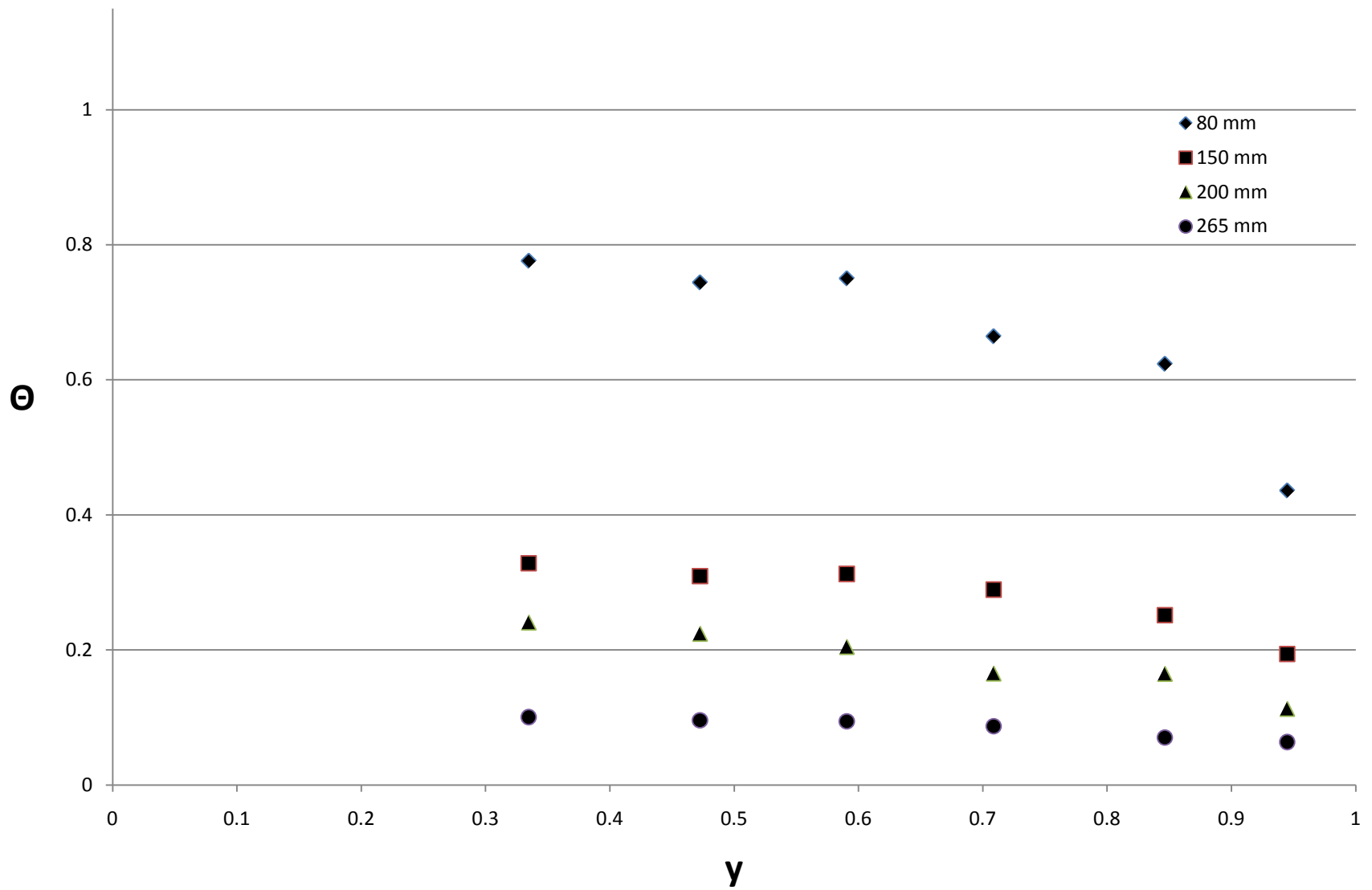
## 2 inch column raschig ring Re 1209



## 2 inch column raschig ring Re 1570

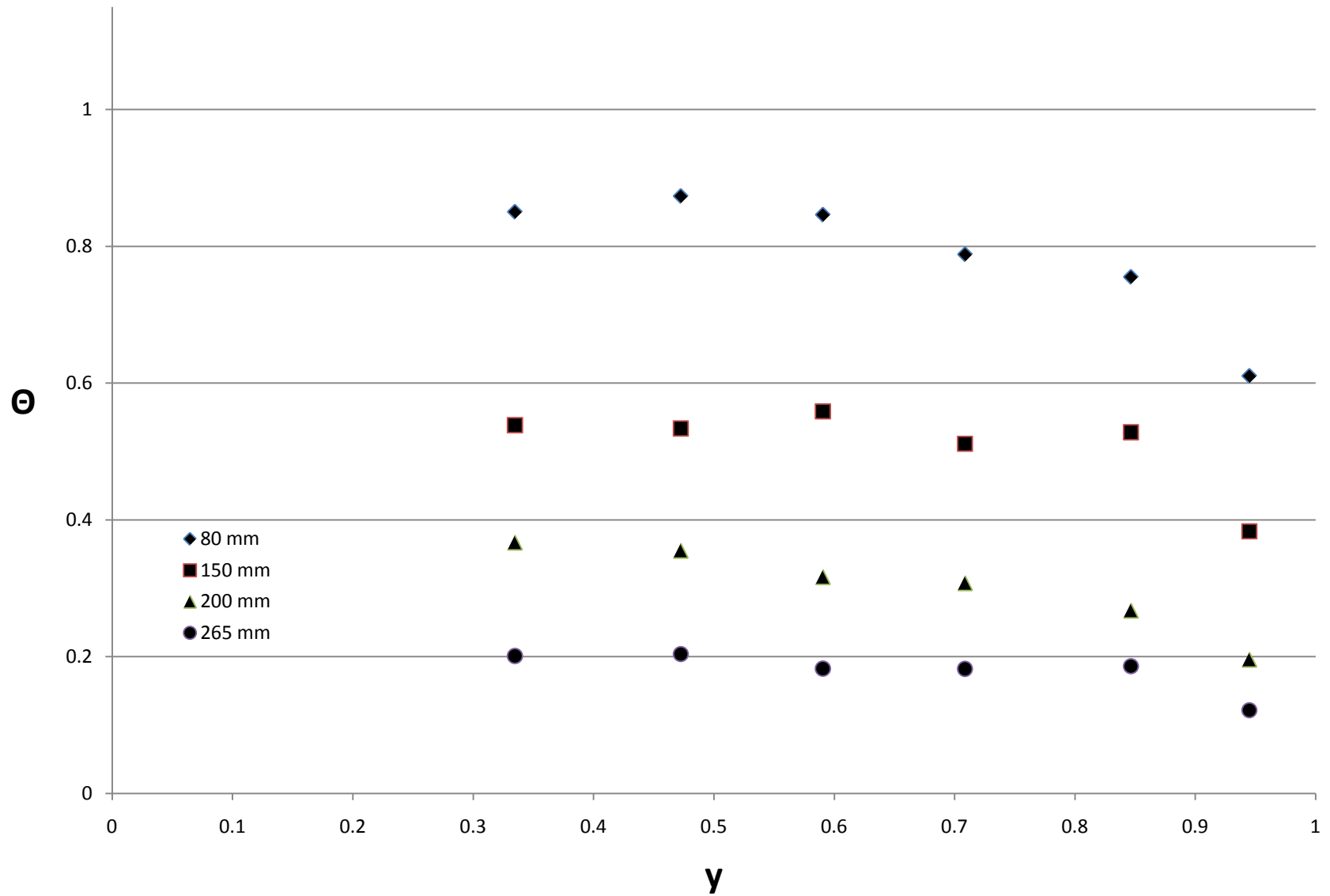


## 2 inch column JM 4-hole cylinders Re 409

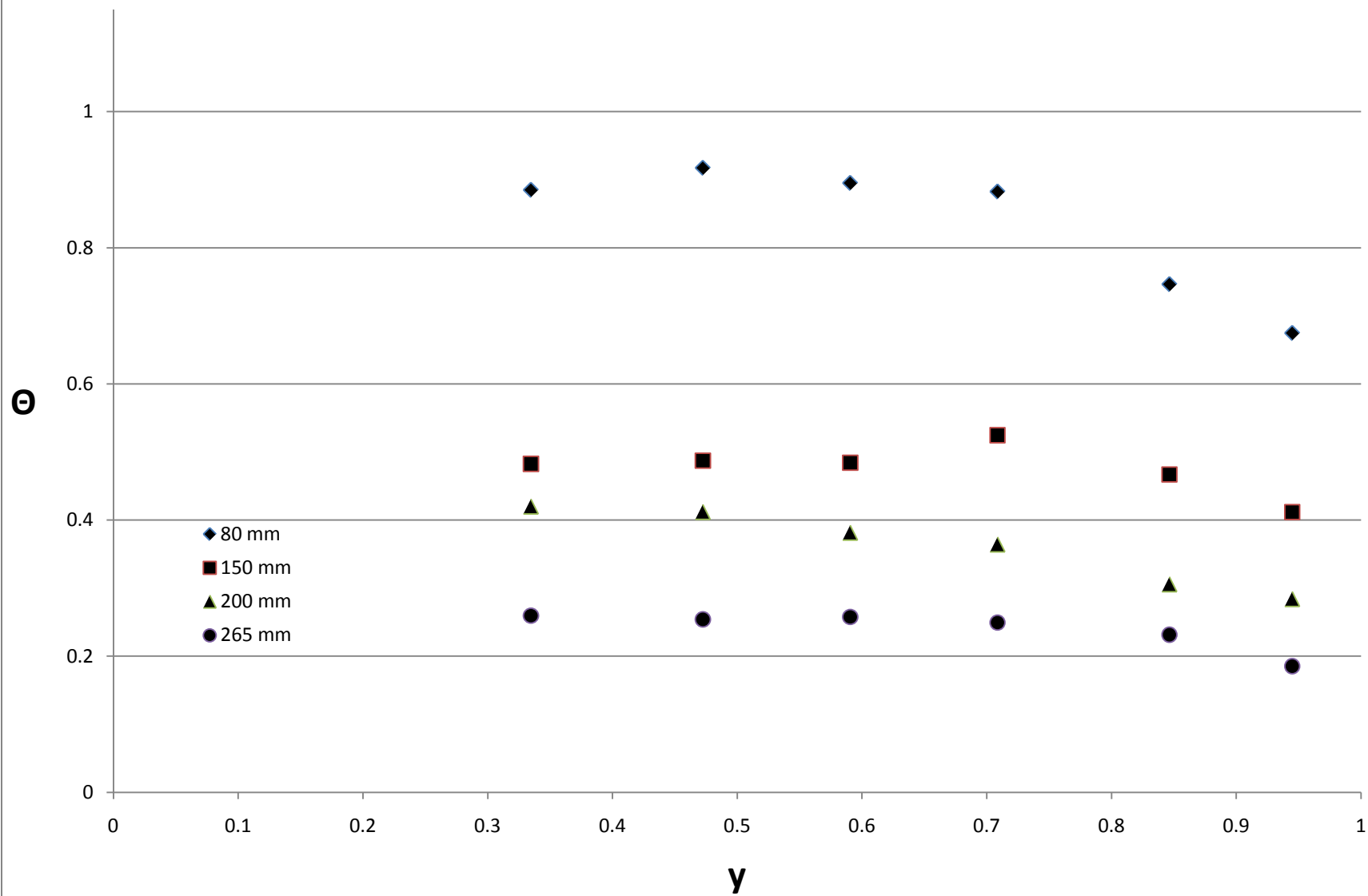




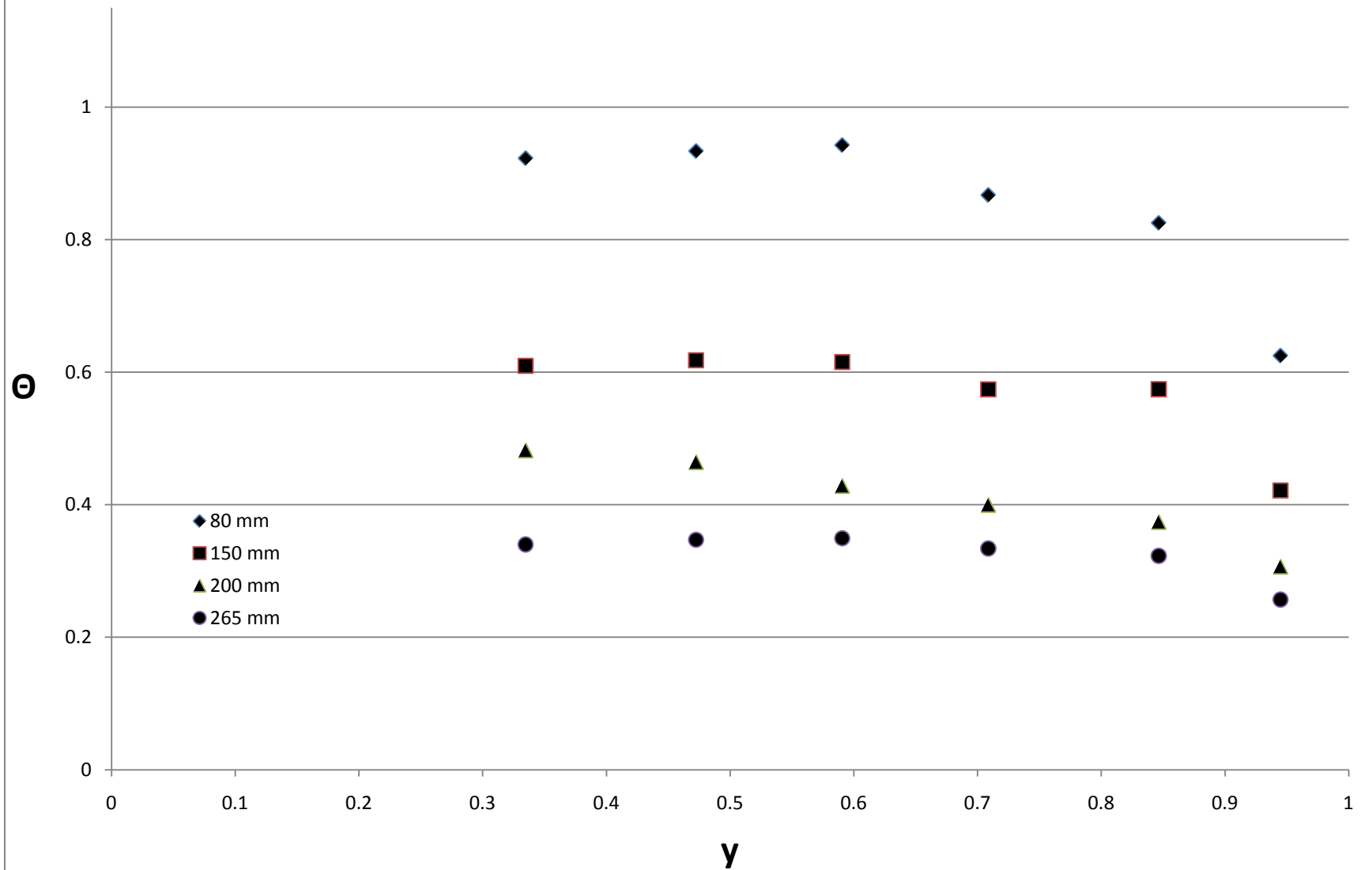
## 2 inch column JM 4-hole cylinders Re 775

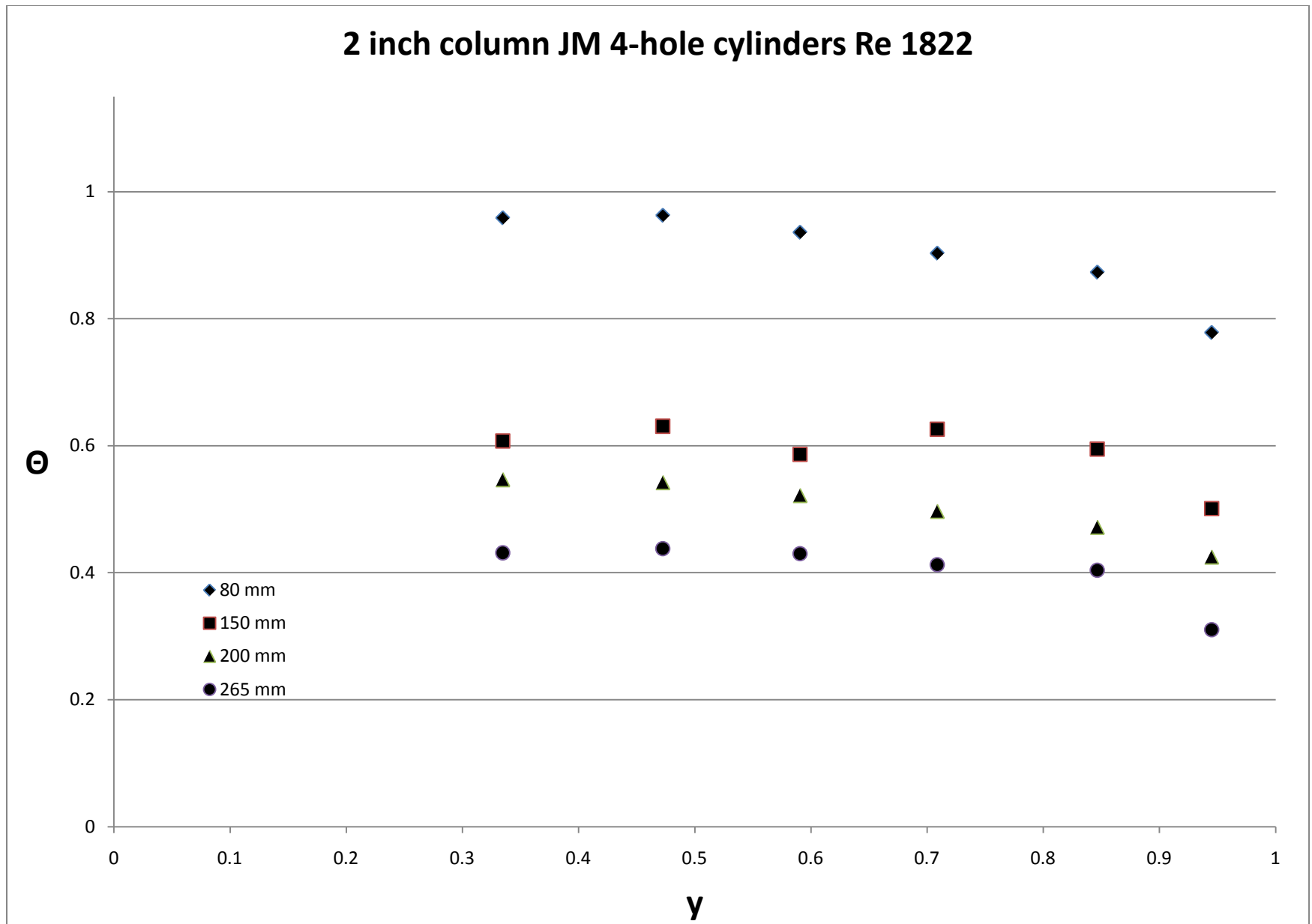


## 2 inch column JM 4-hole cylinders Re 1052

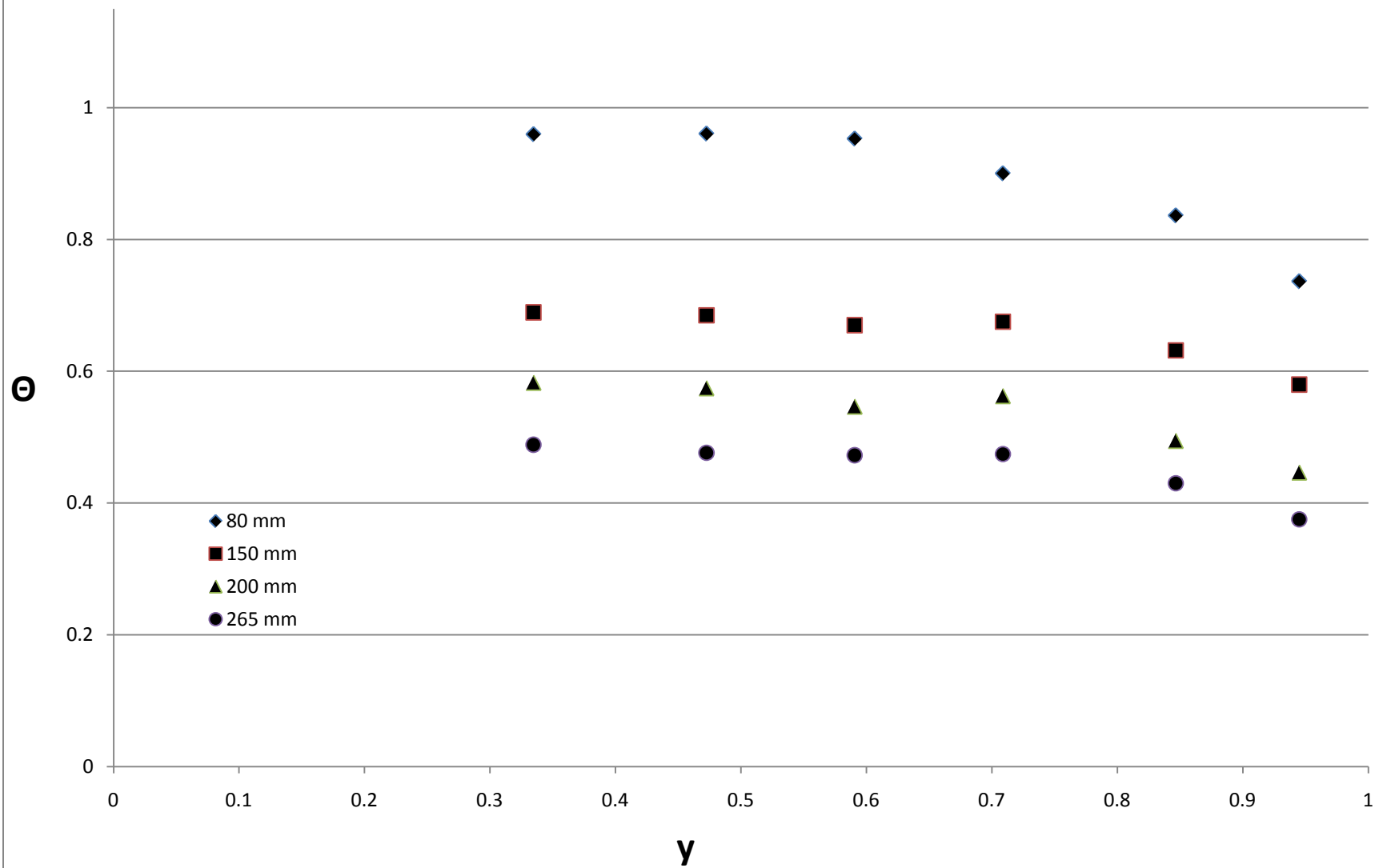


## 2 inch column JM 4-hole cylinders Re 1412

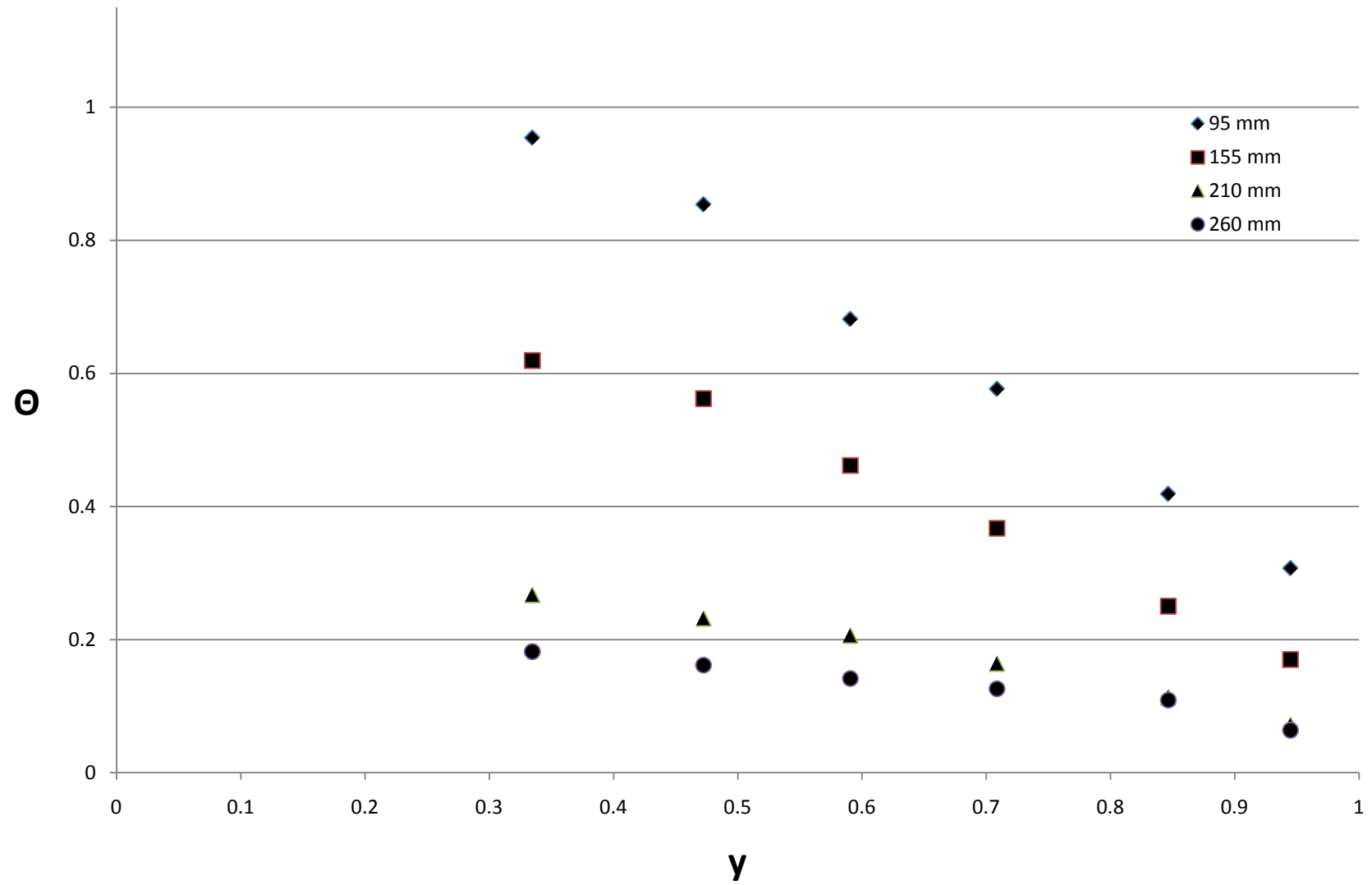




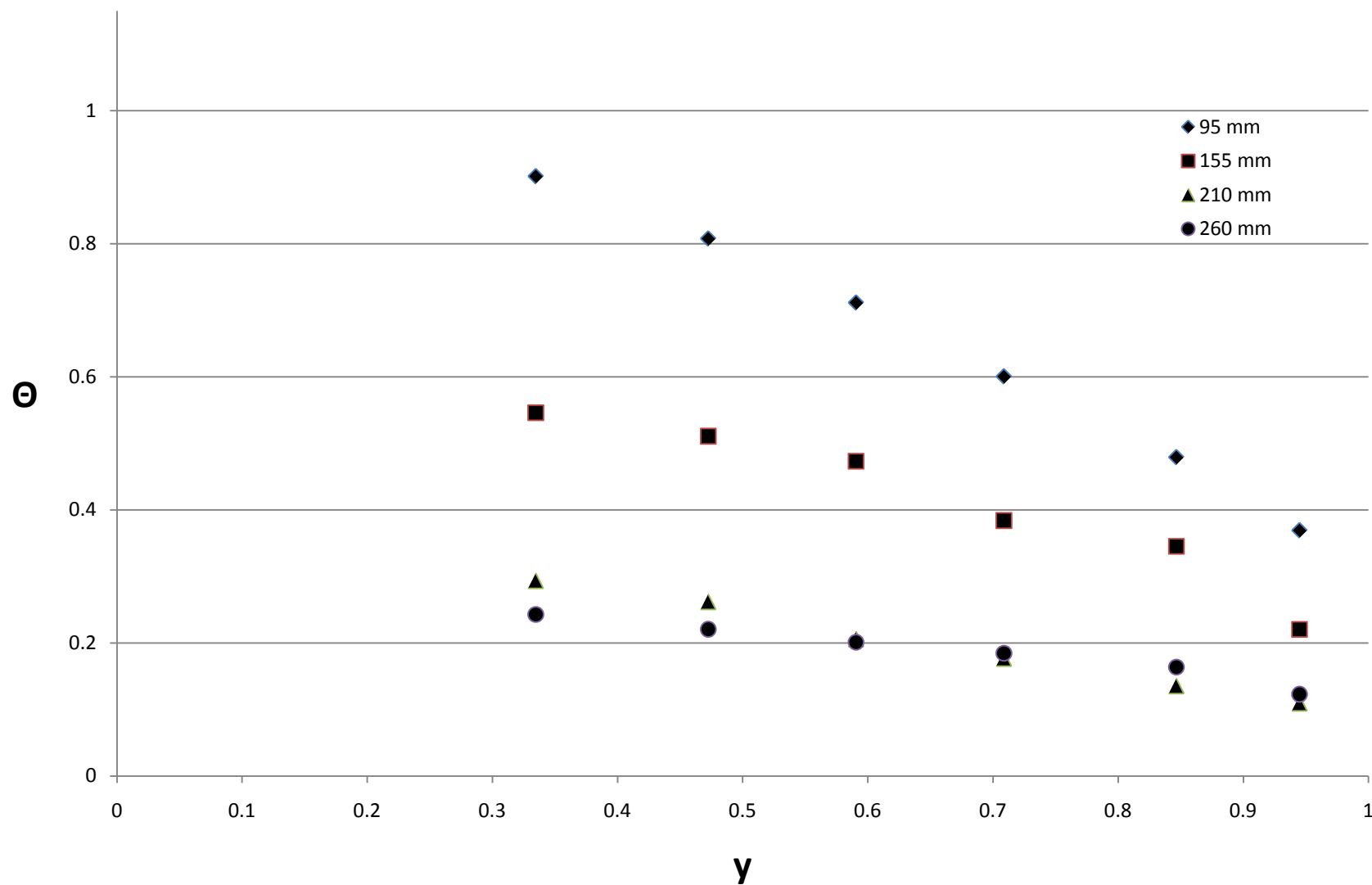
## 2 inch column JM 4-hole cylinders Re 2275



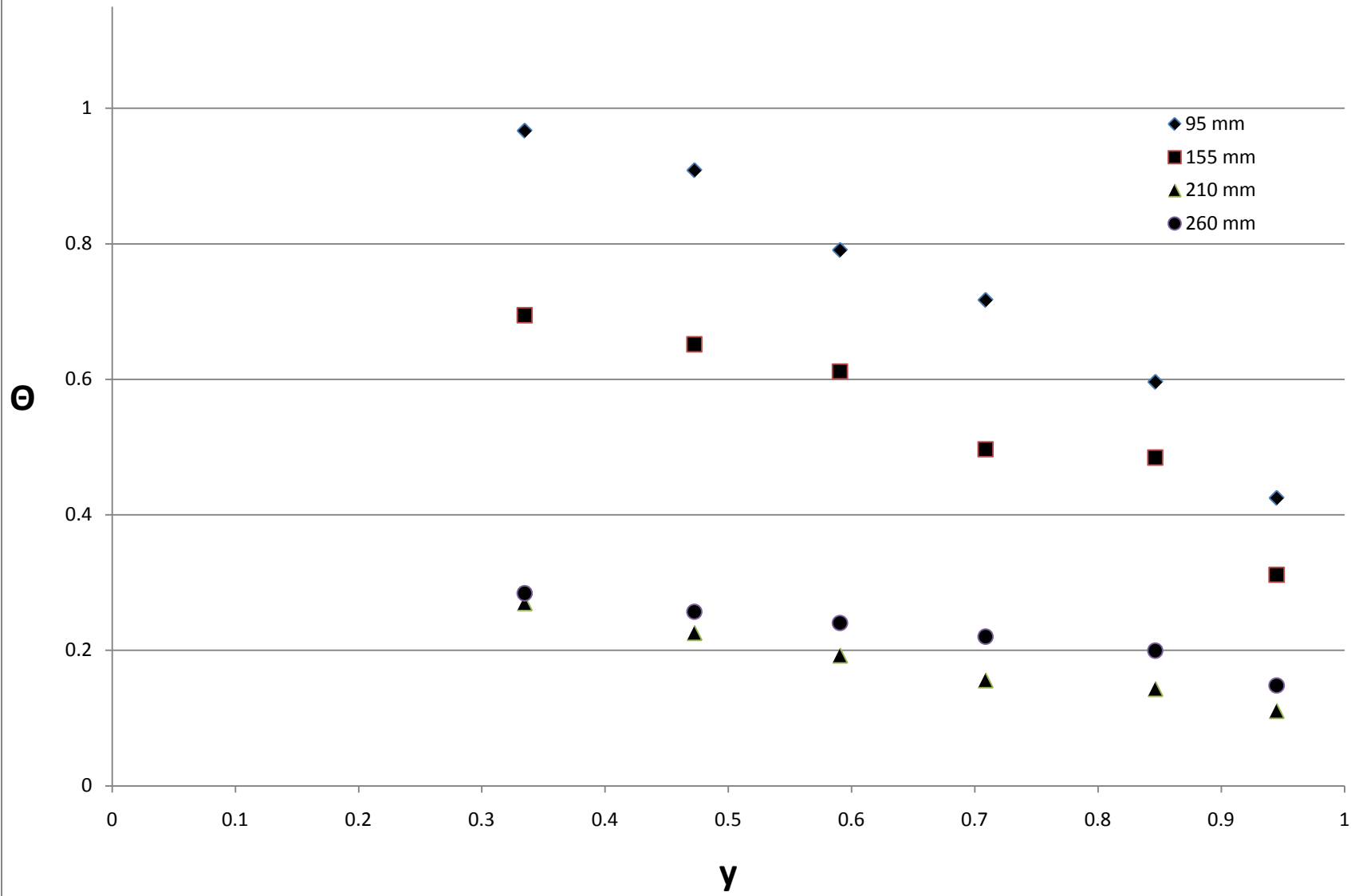
## 2 inch column monoliths Re 170



## 2 inch column monoliths Re 323

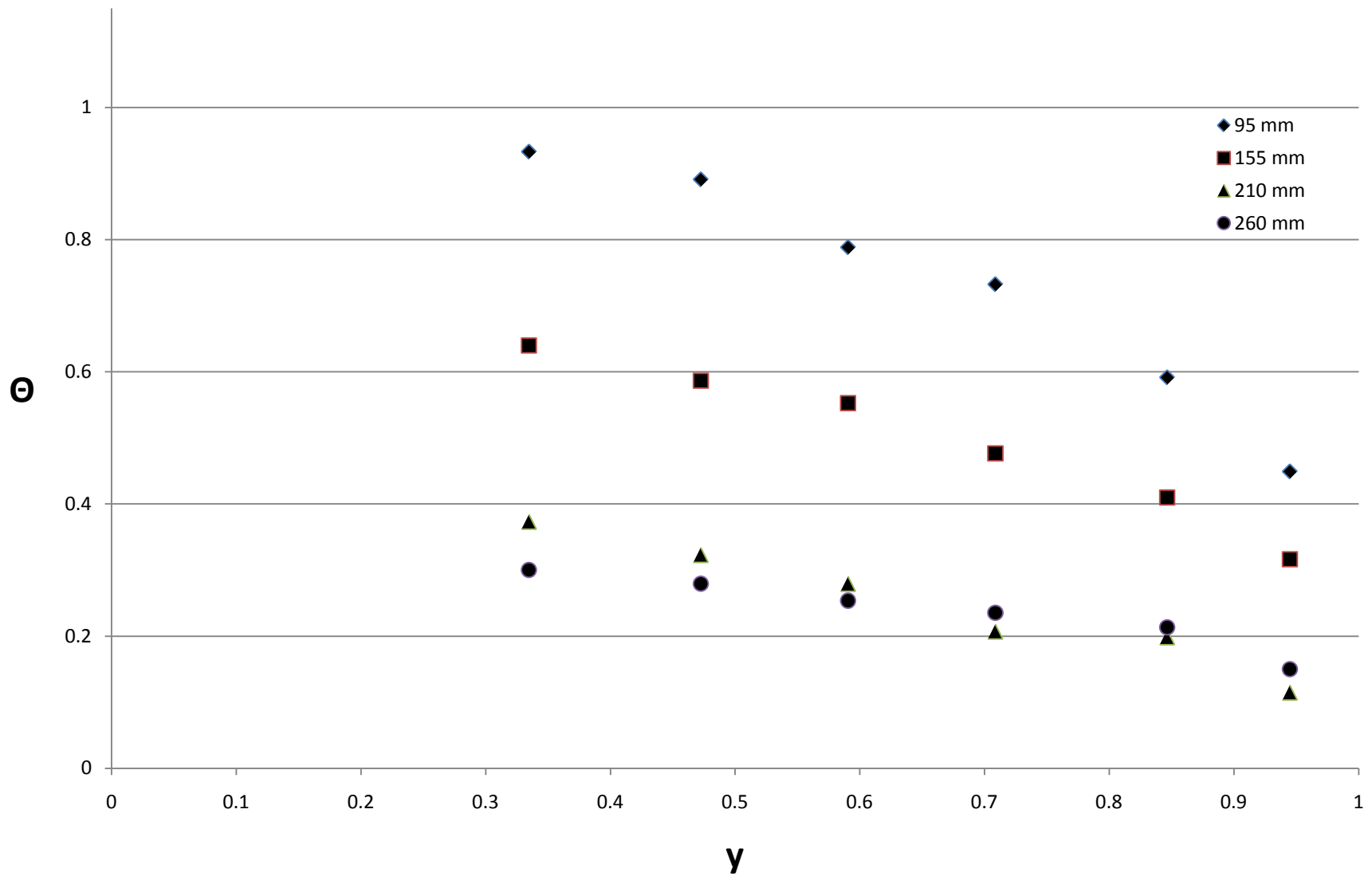


## 2 inch column monoliths Re 439

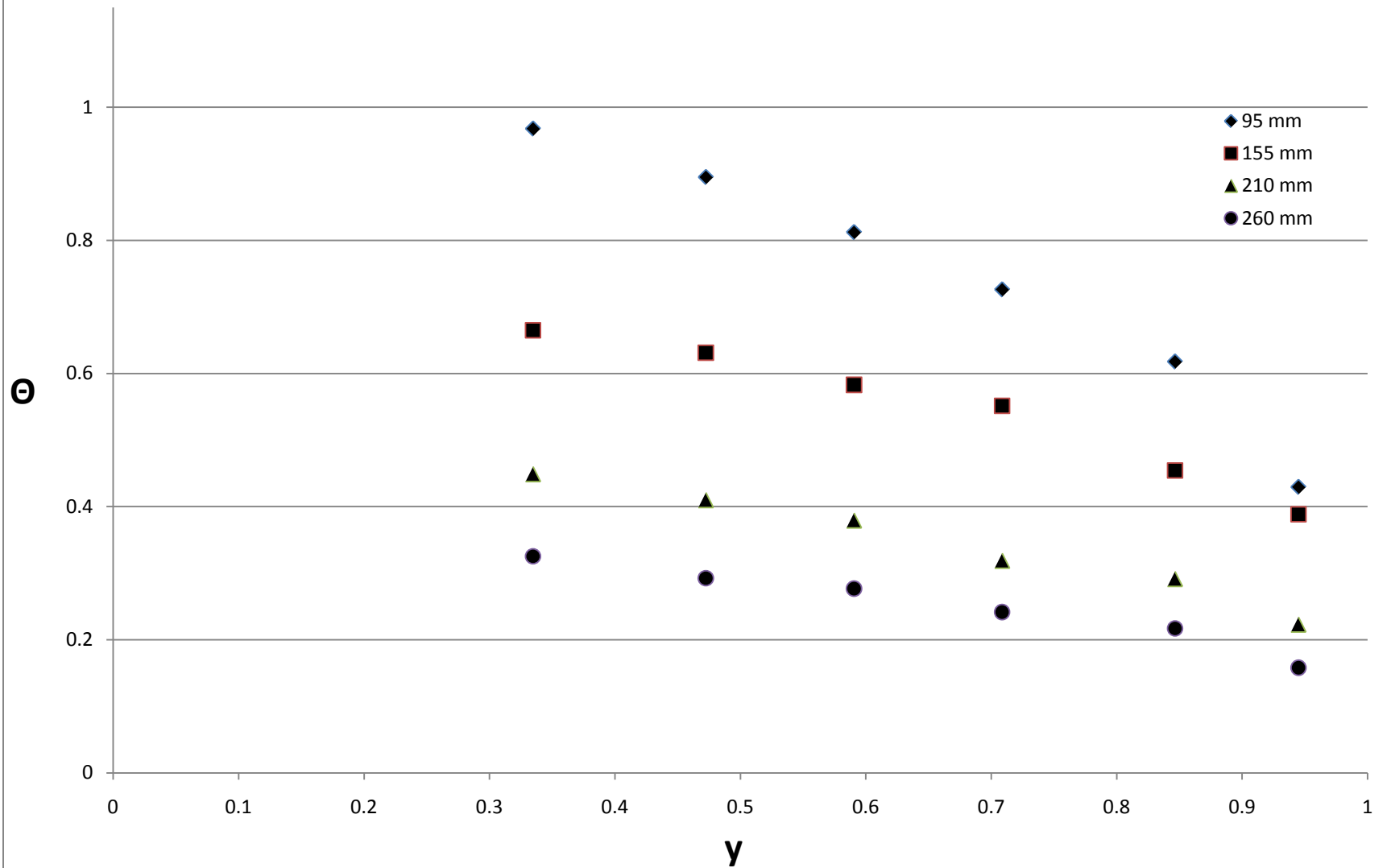




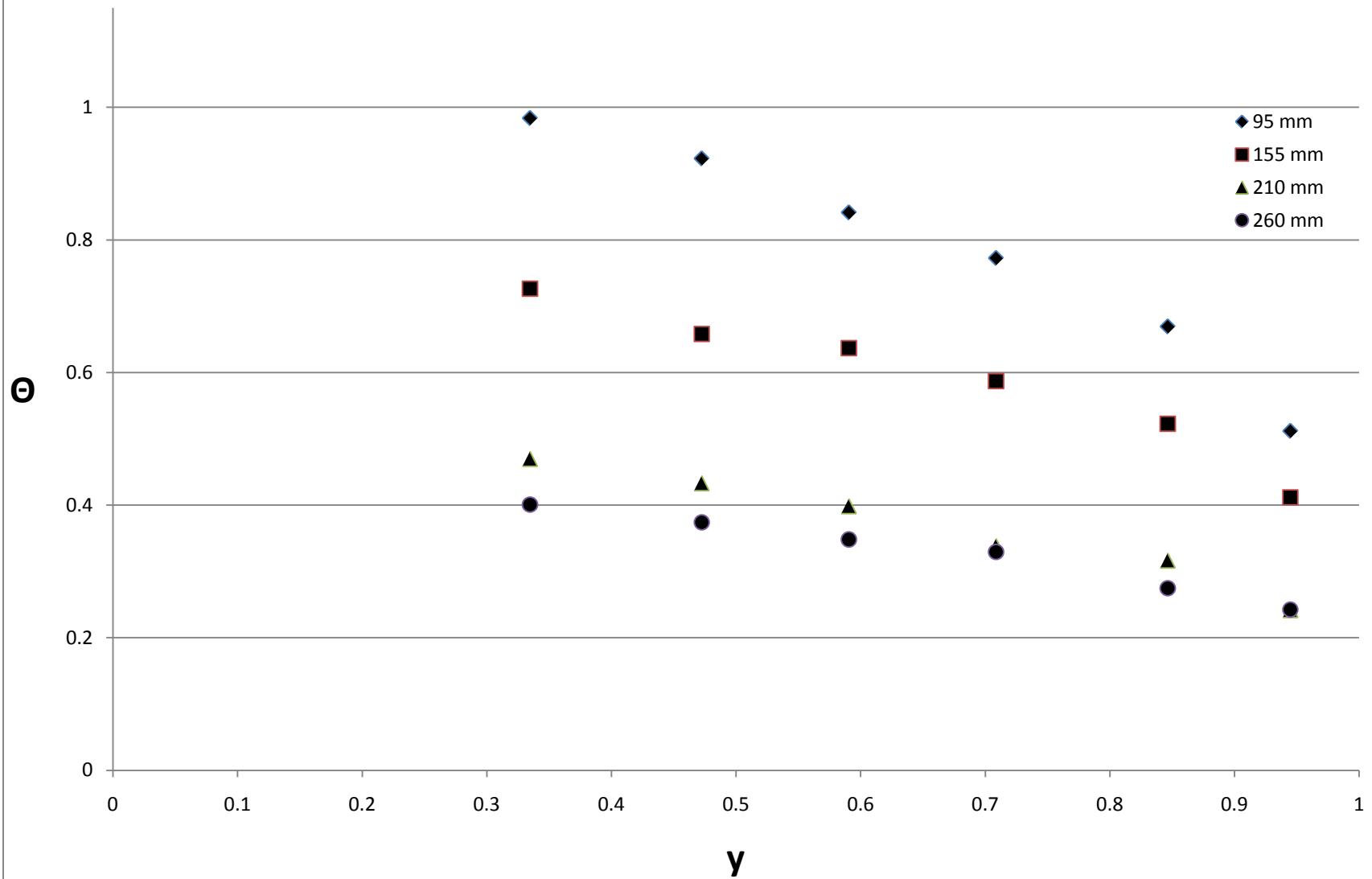
## 2 inch column monoliths Re 589



## 2 inch column monoliths Re 760

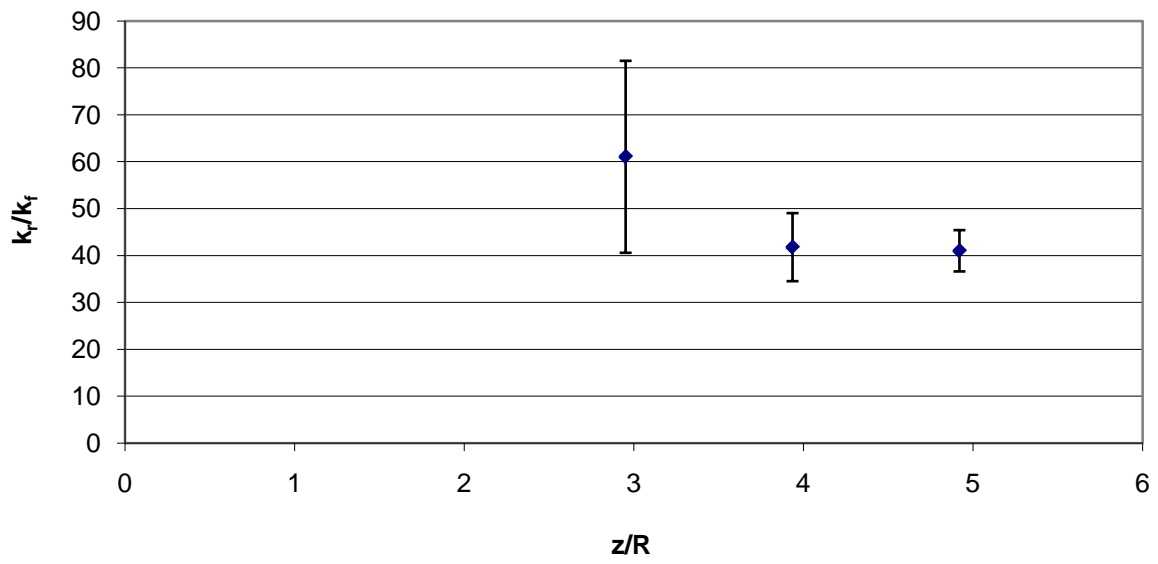


## 2 inch column monoliths Re 948

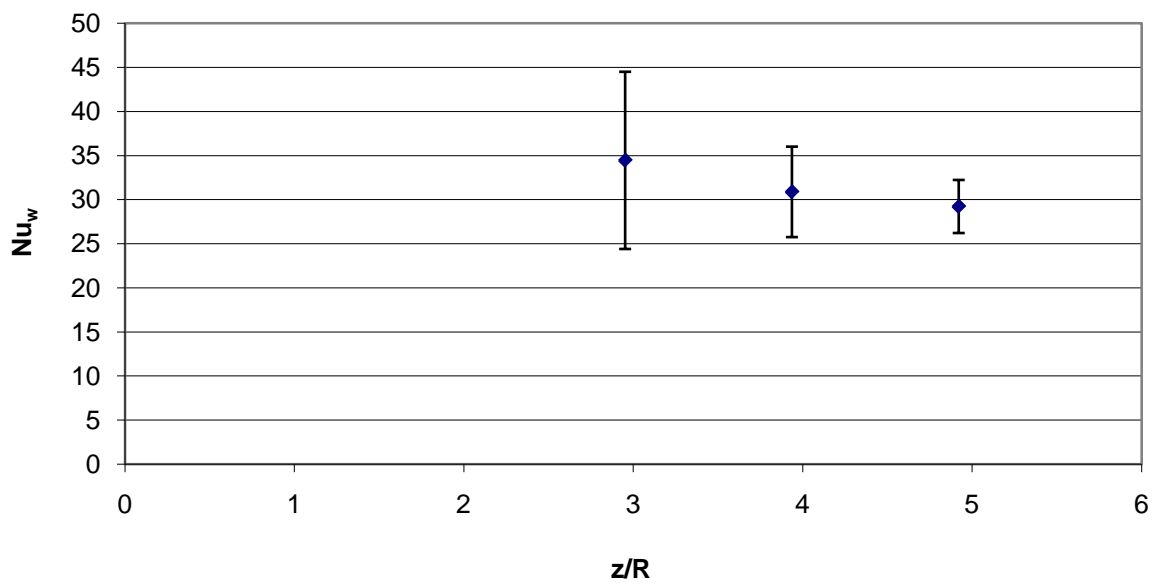


## Appendix D: $k_r/k_f$ and $Nu_w$ Height by Height Analysis

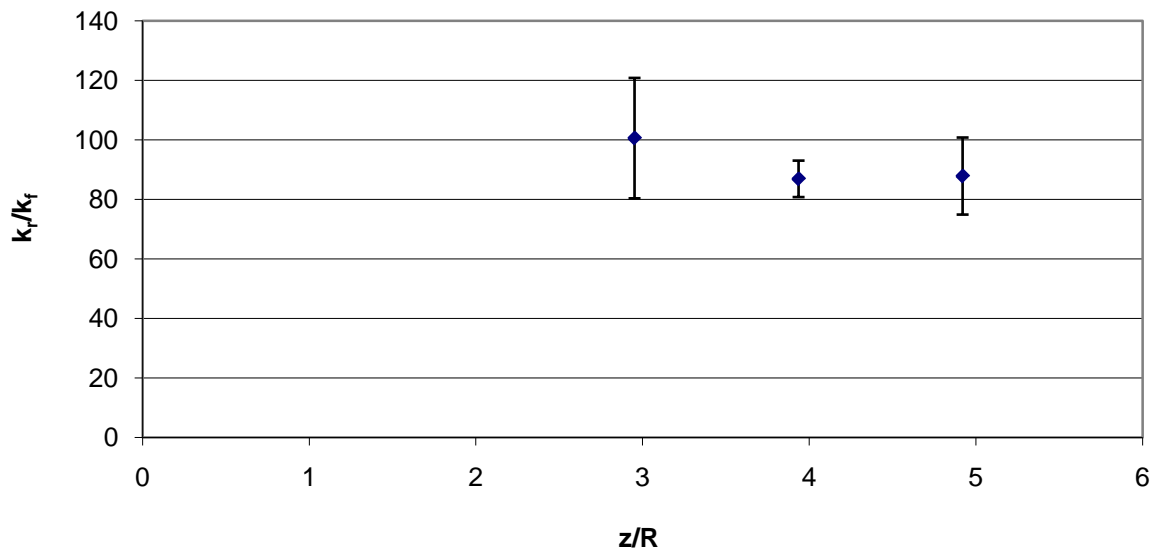
4 inch column Raschig Ring Re 258



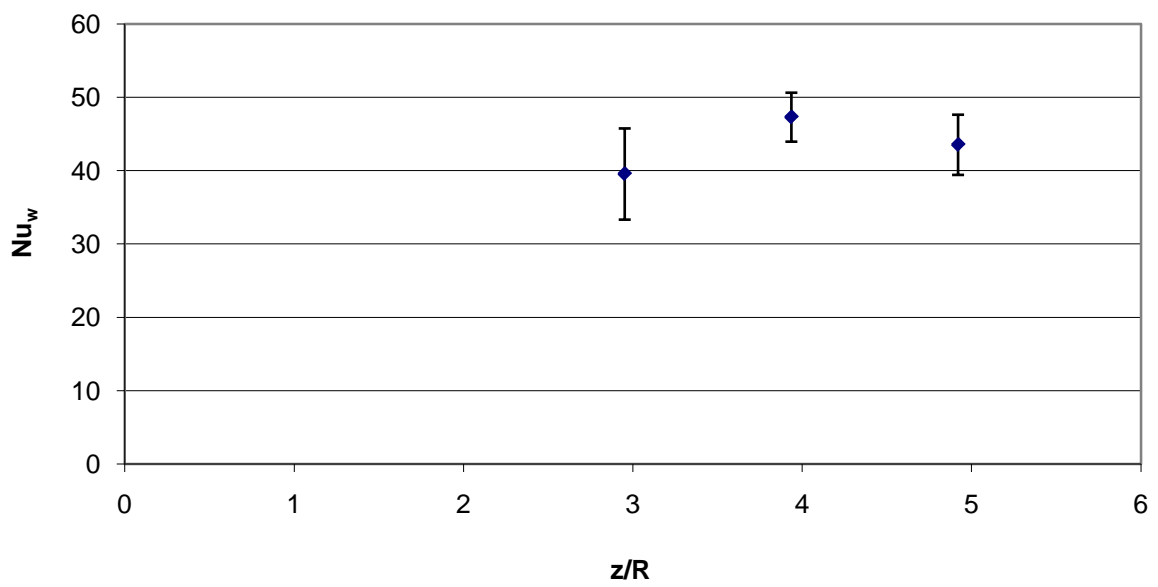
4 inch column Raschig Ring Re 258



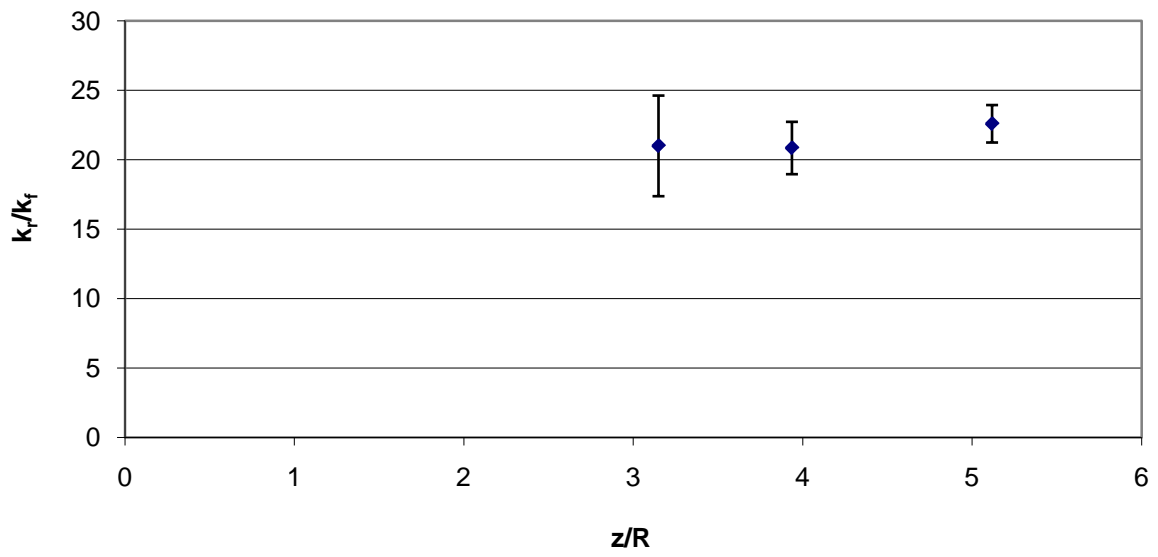
**4 inch column JM 4-hole cylinders Re 584**



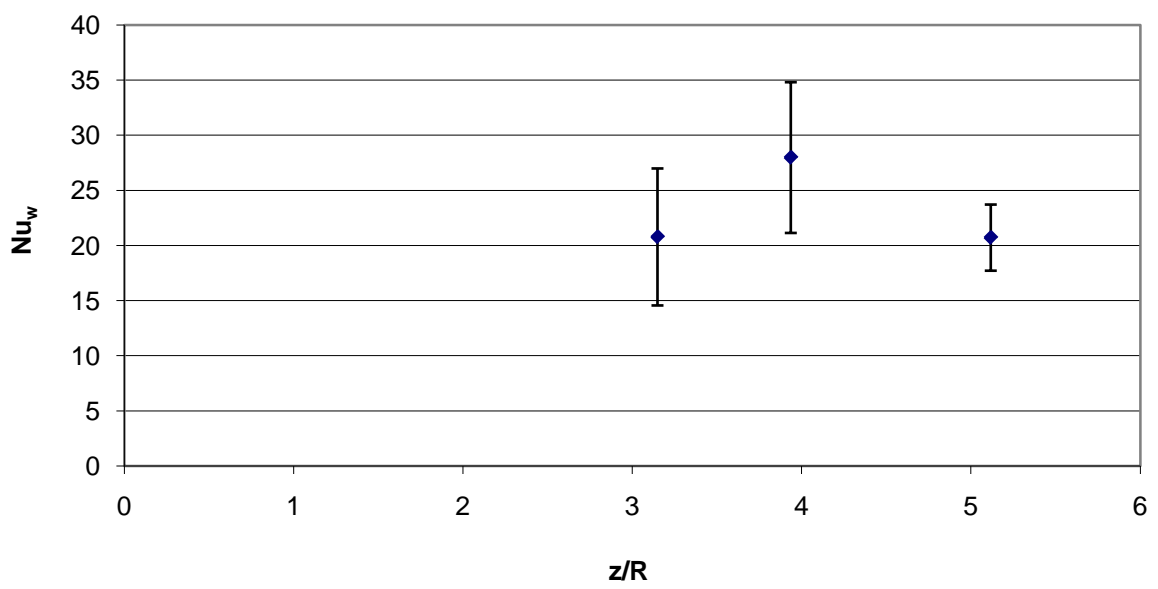
**4 inch column JM 4-hole cylinders Re 584**



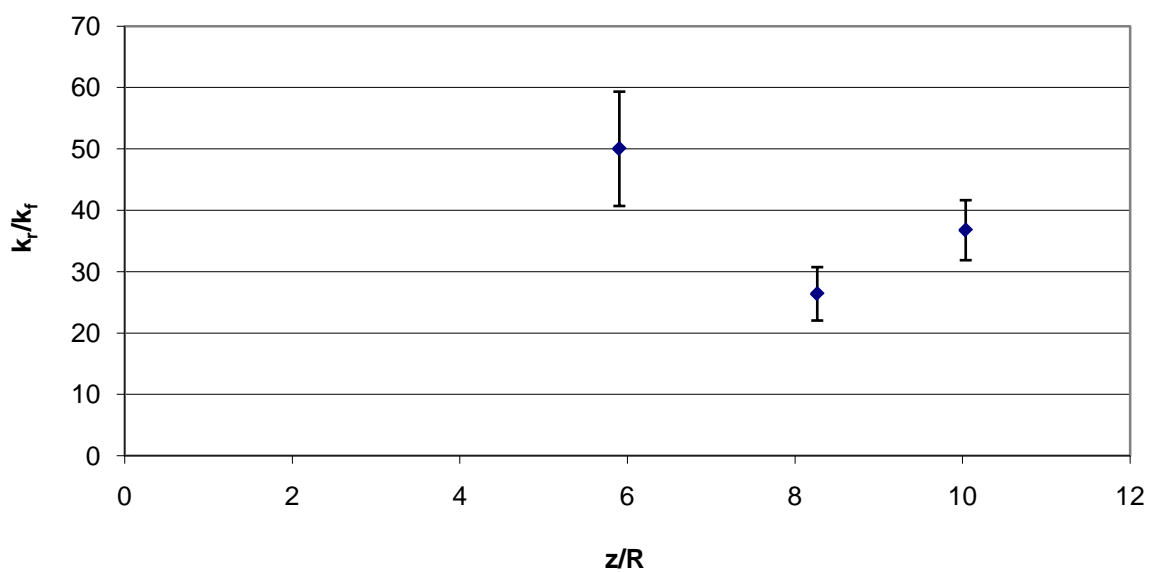
**4 inch column Monoliths Re 95**



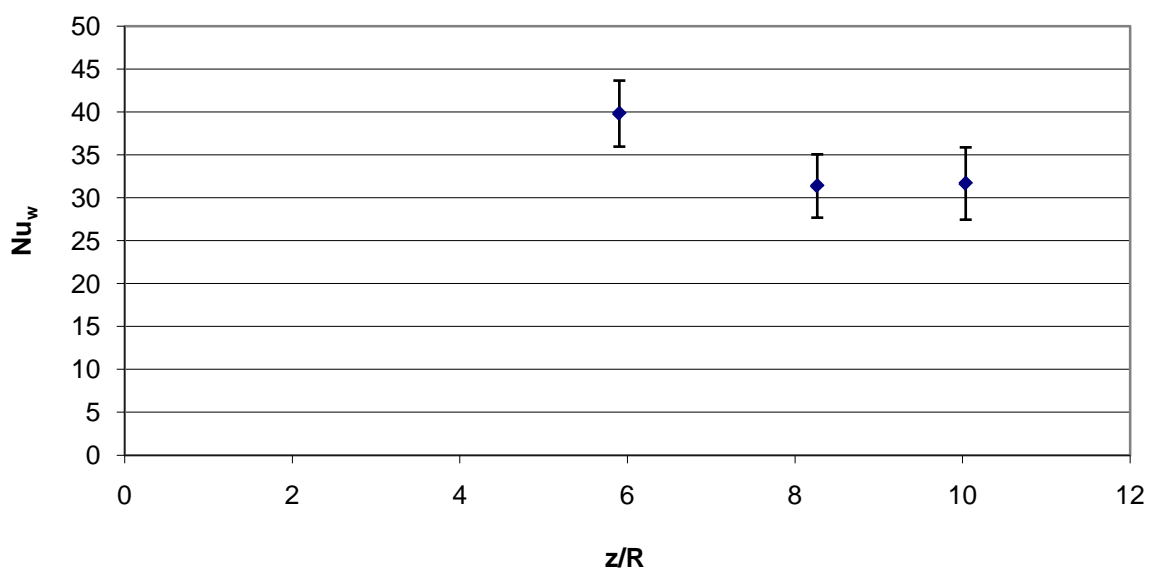
**4 inch column Monoliths Re 95**



**2 inch column Raschig Ring Re 350**

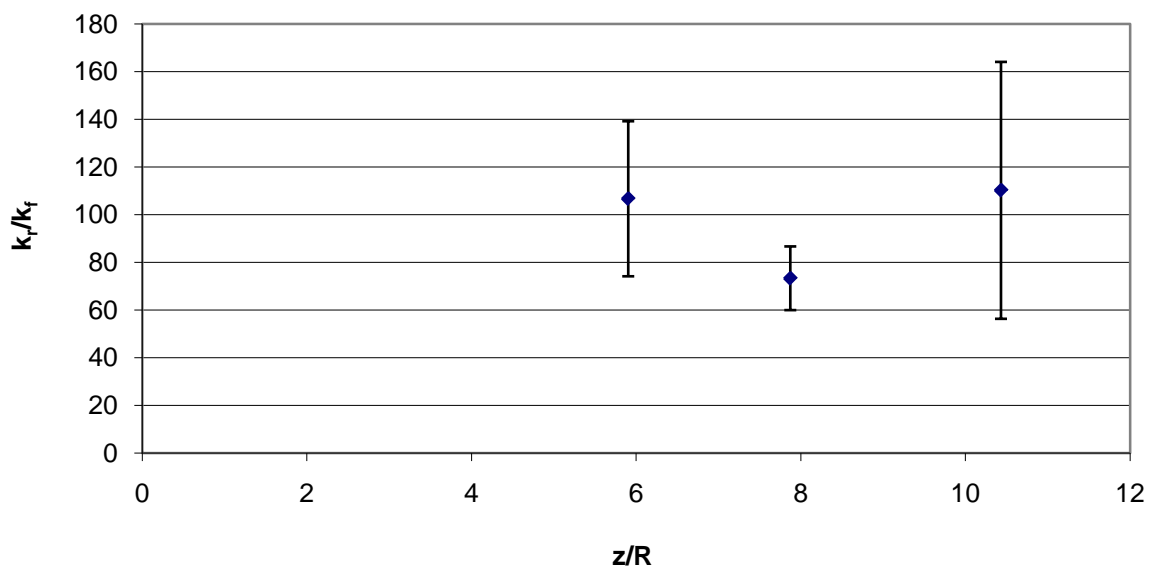


**2 inch column Raschig Ring Re 350**

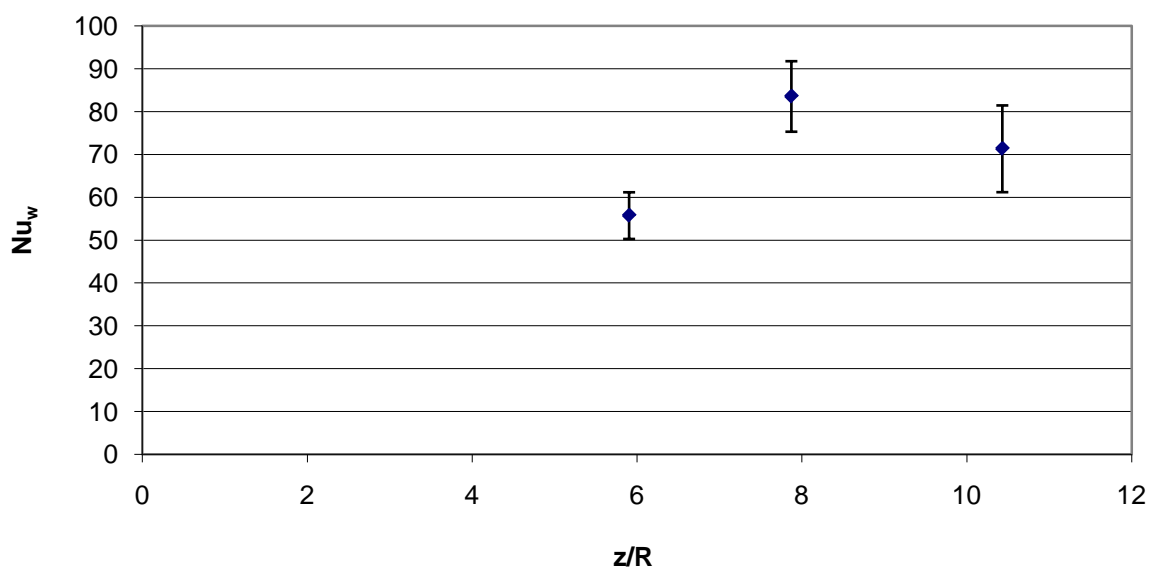


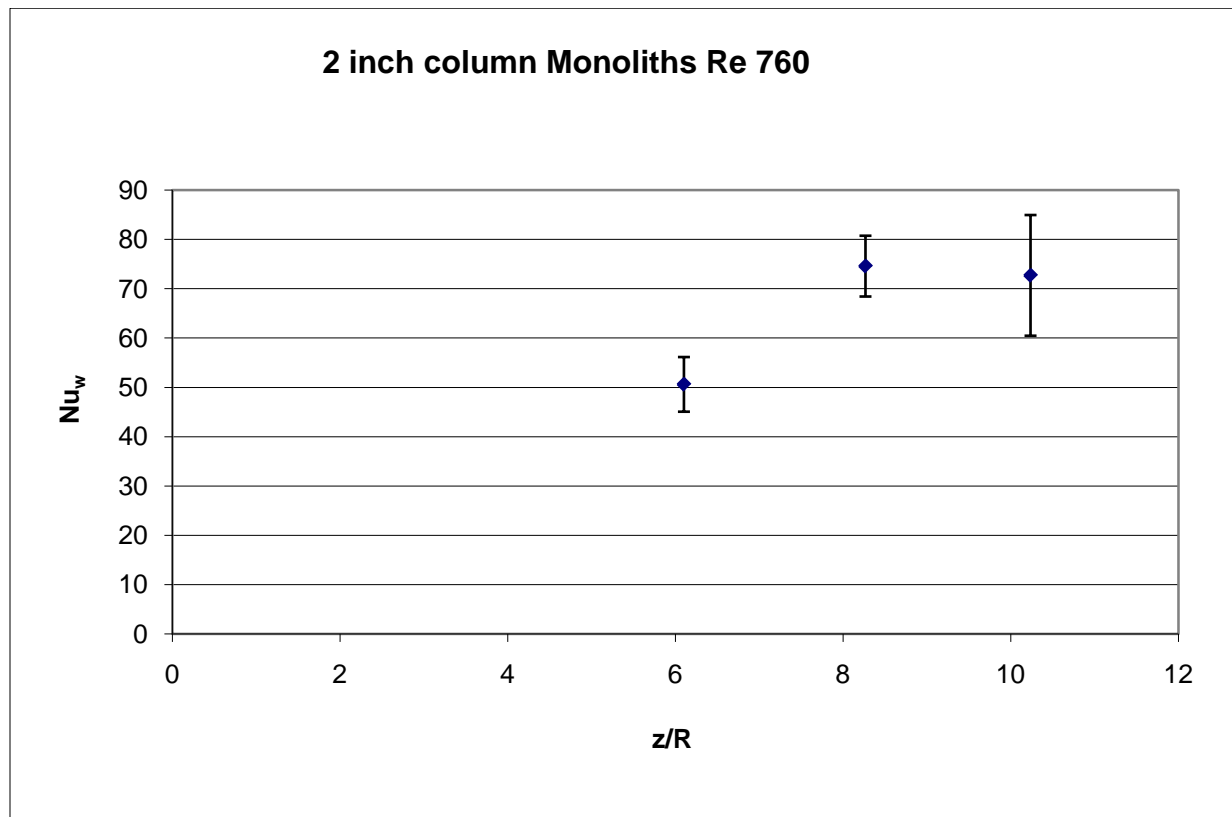
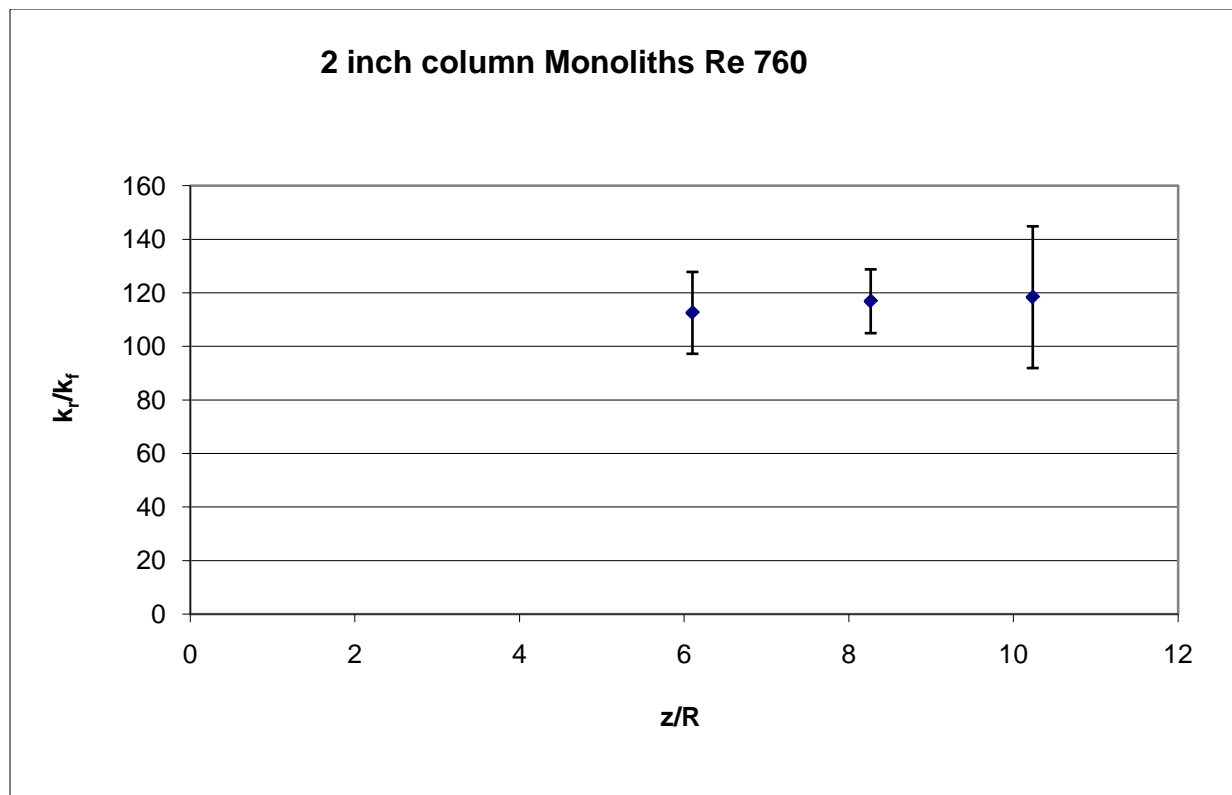


**2 inch column JM 4-hole cylinders Re 775**



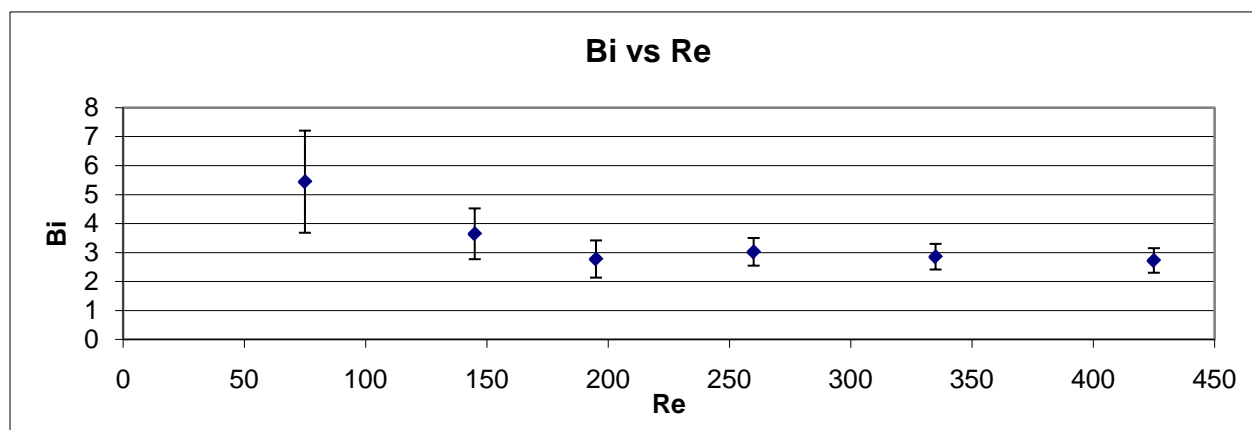
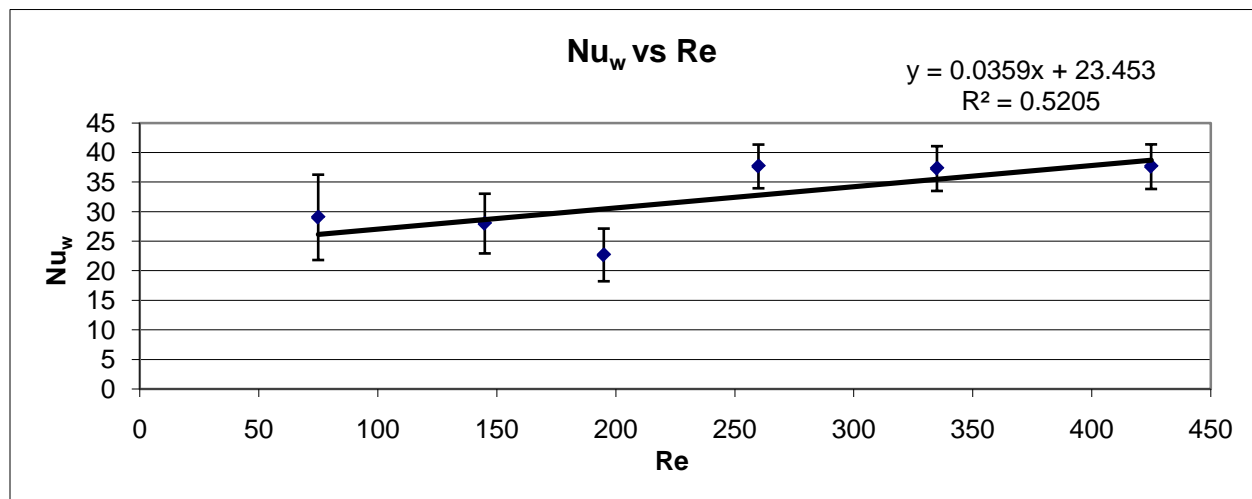
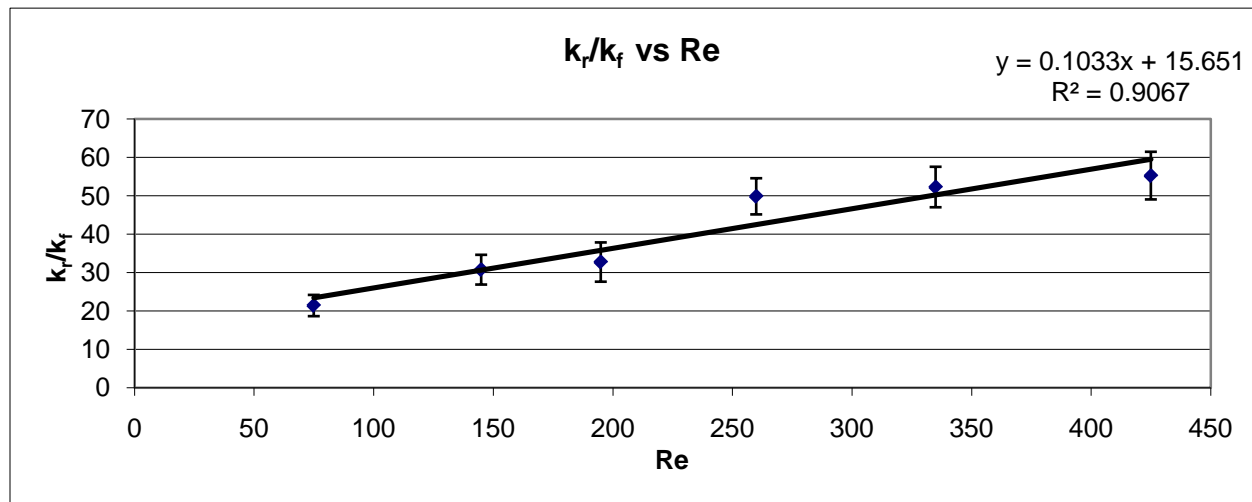
**2 inch column JM 4-hole cylinders Re 775**

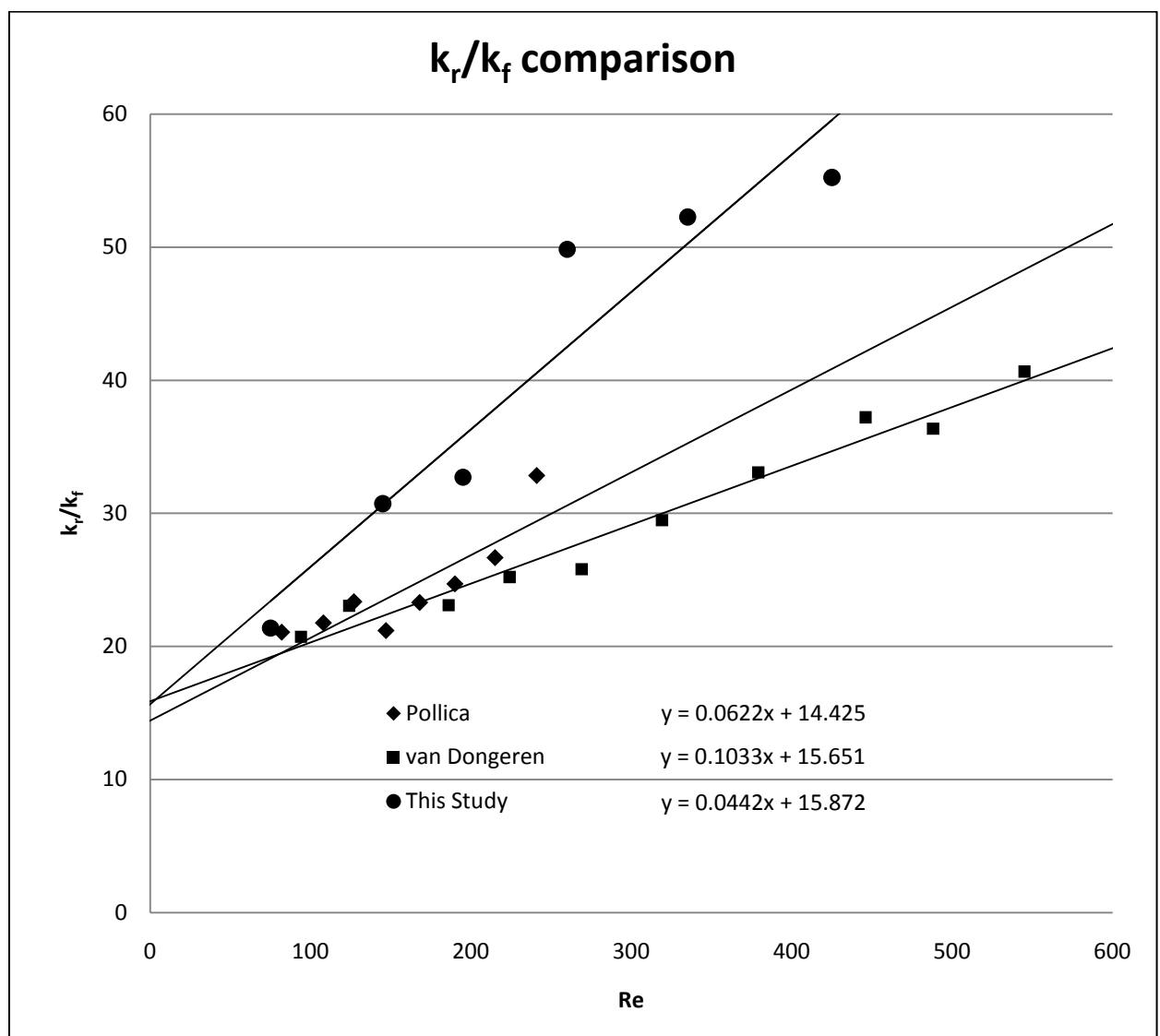
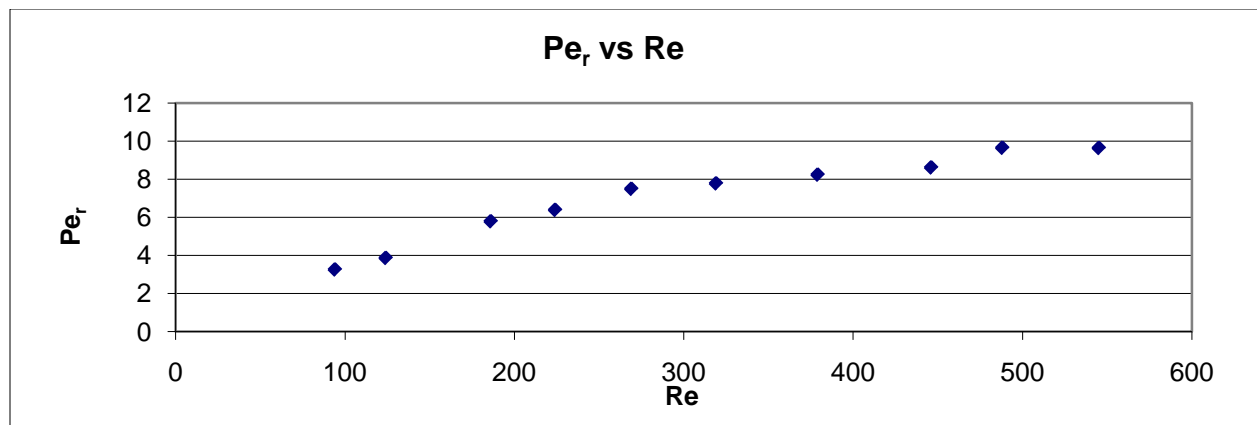


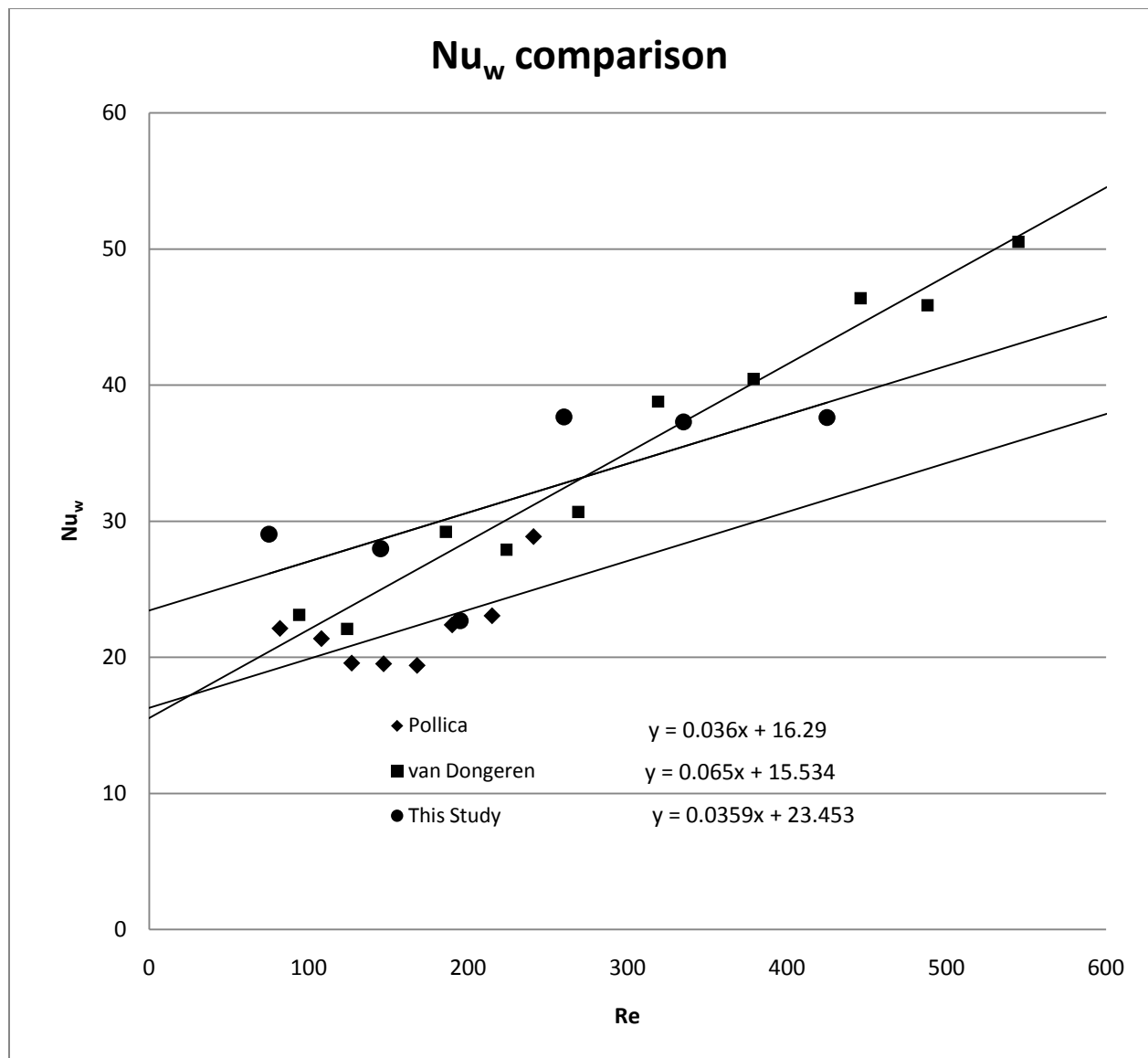


## Appendix E: $k_r/k_f$ , $Nu_w$ , $Bi$ , and $Pe_r$ for all Packings and Columns

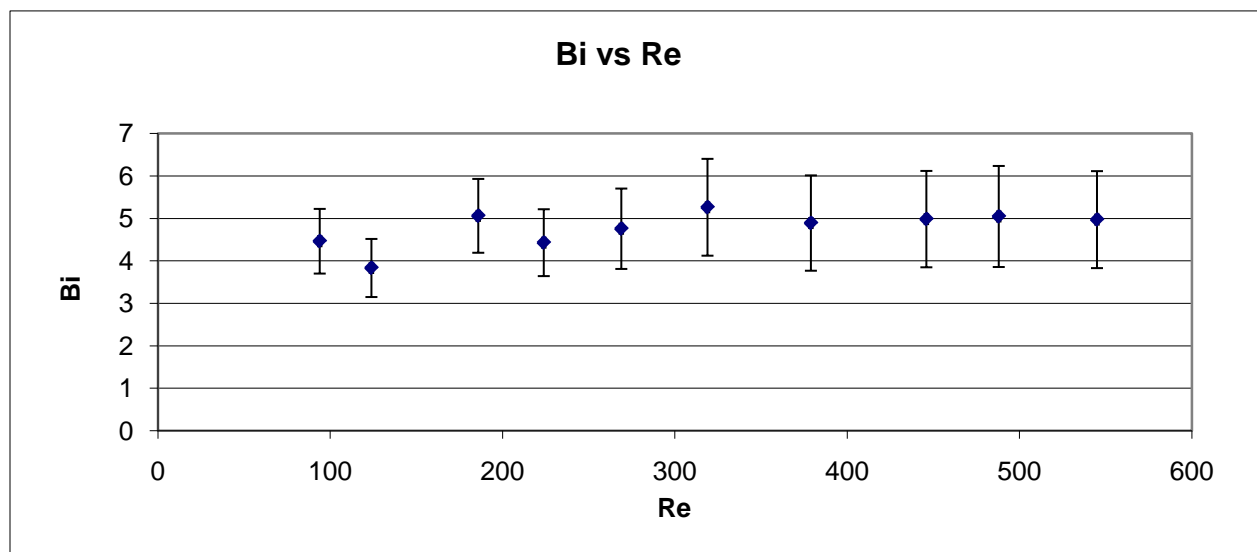
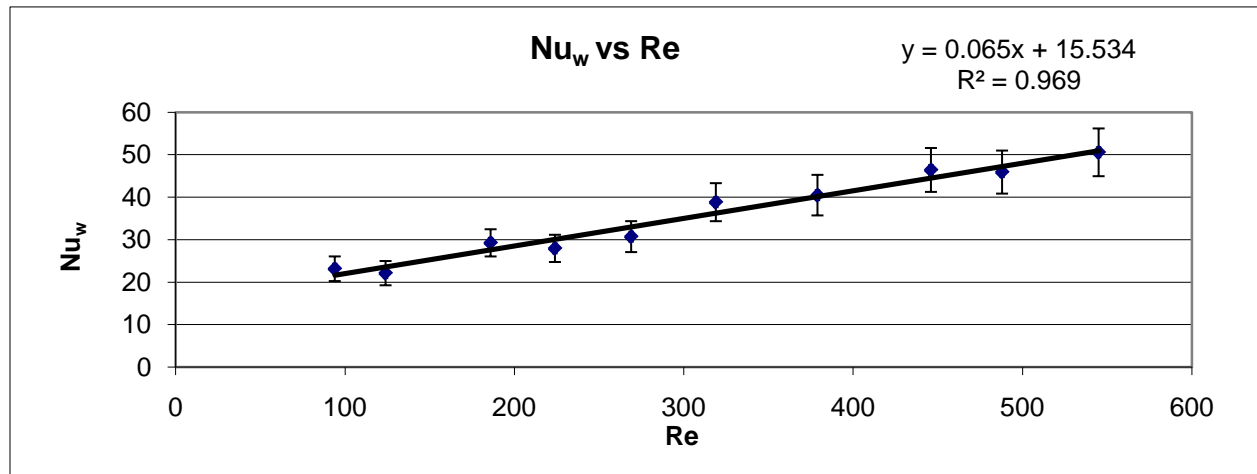
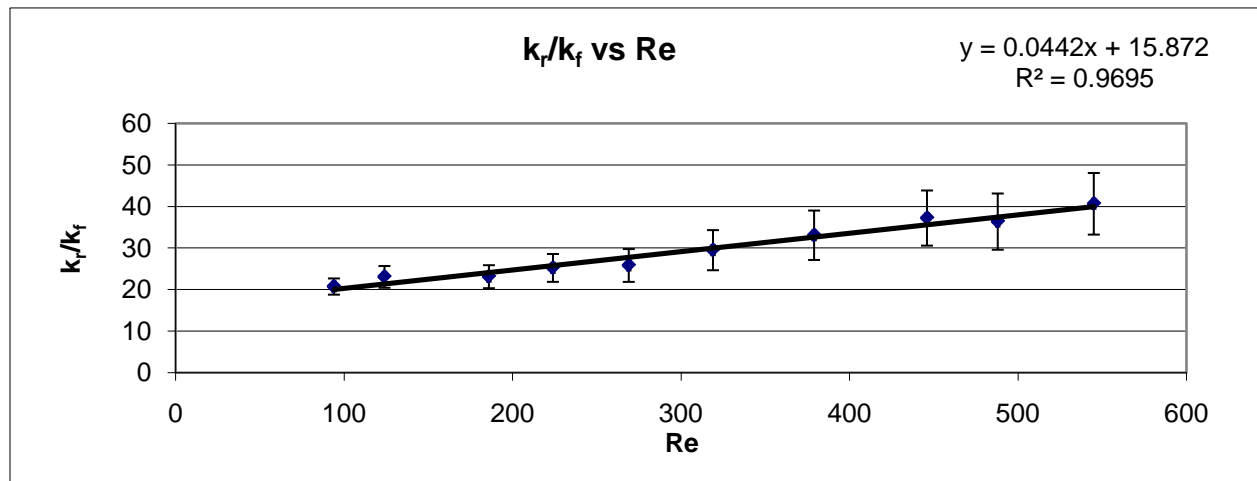
#### 4 inch column Spheres

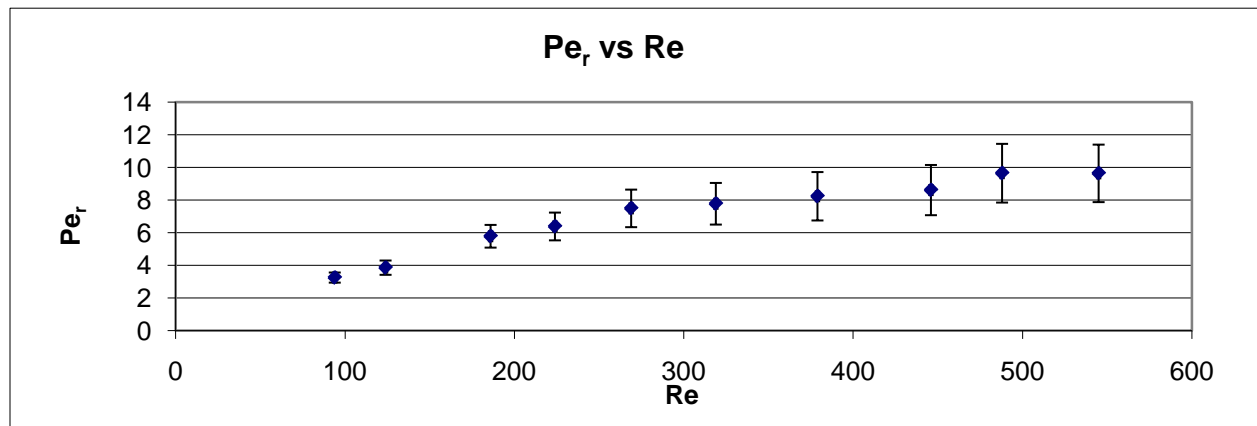






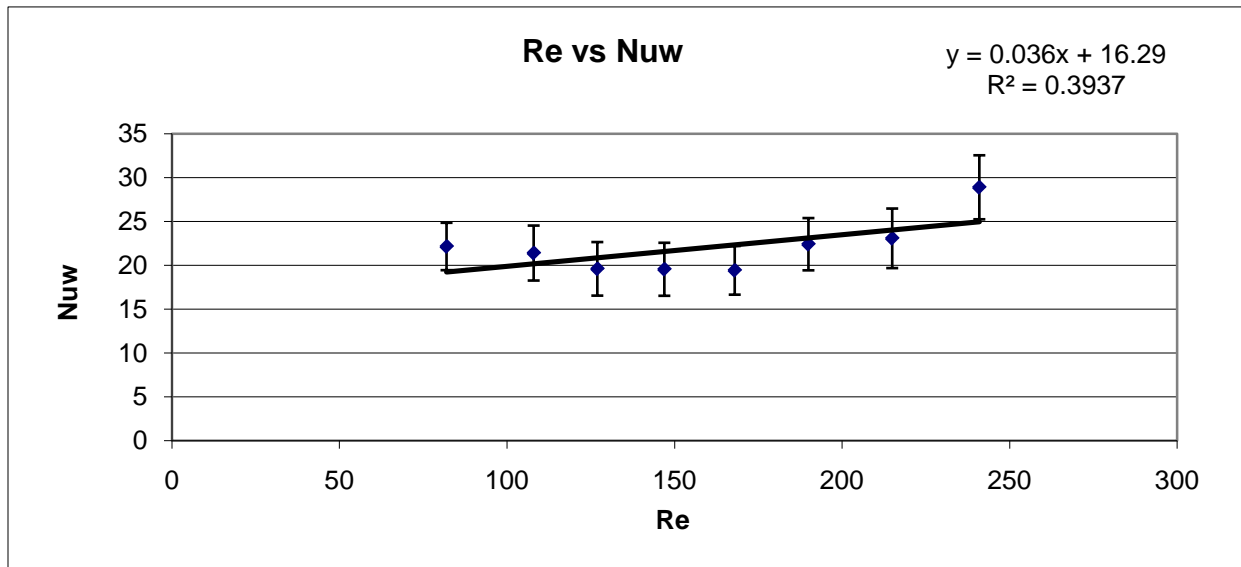
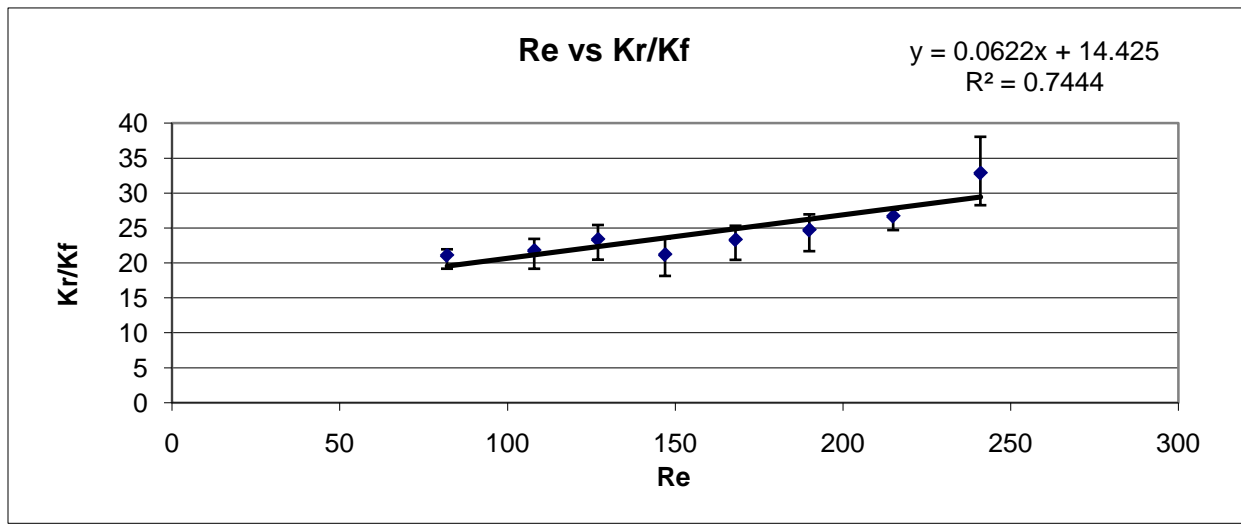
#### 4 inch column Spheres (Van Dongeren)



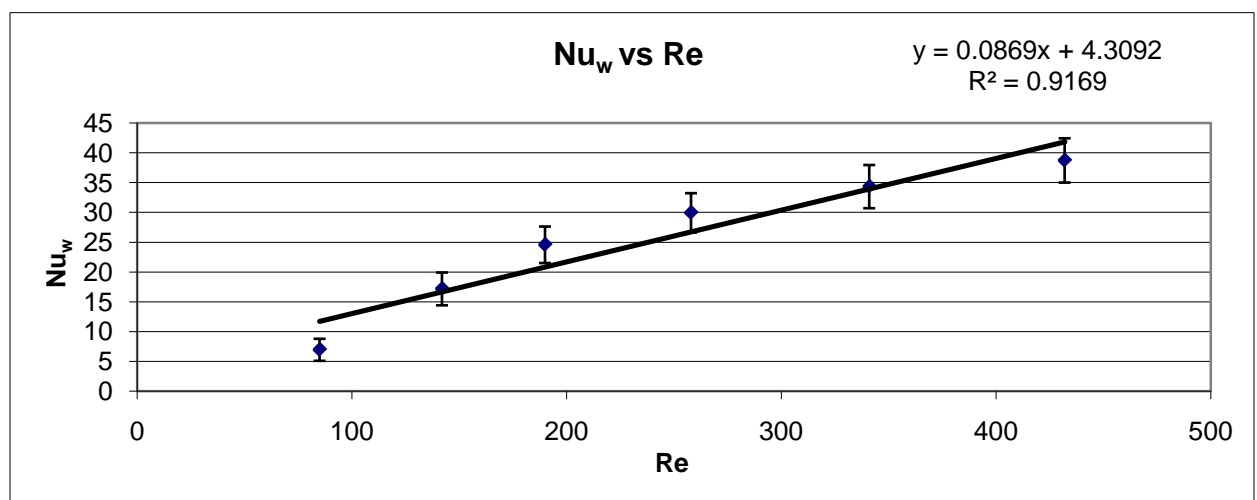
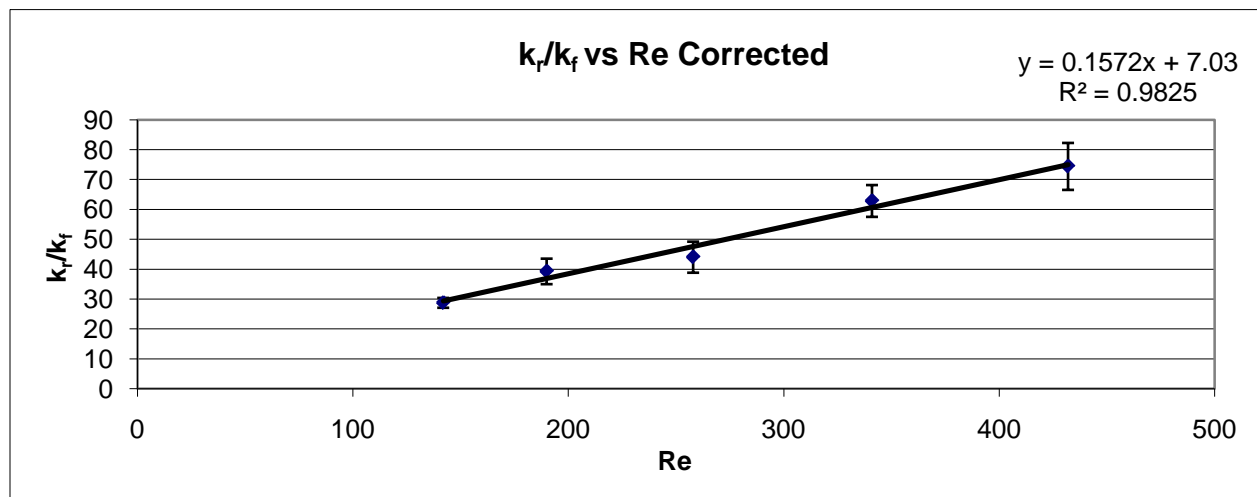
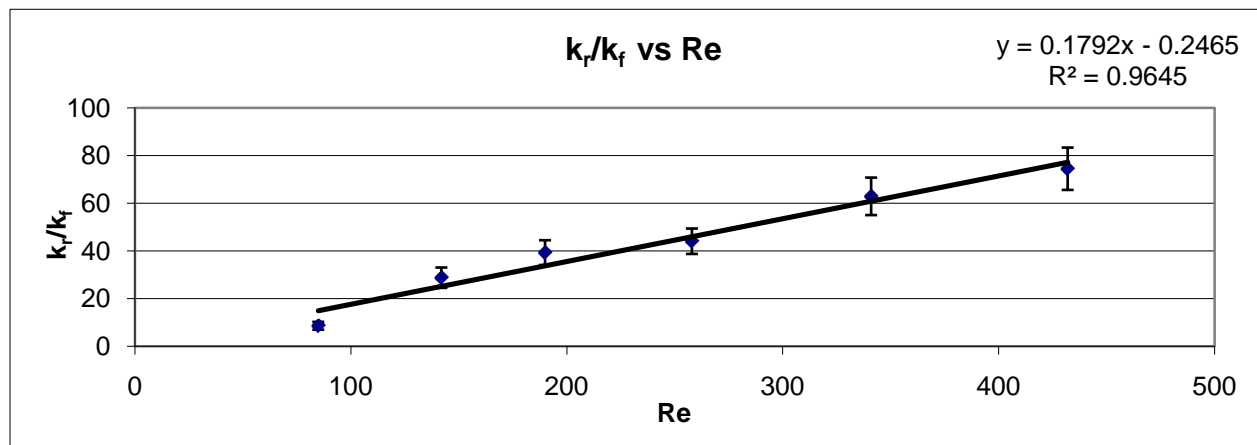


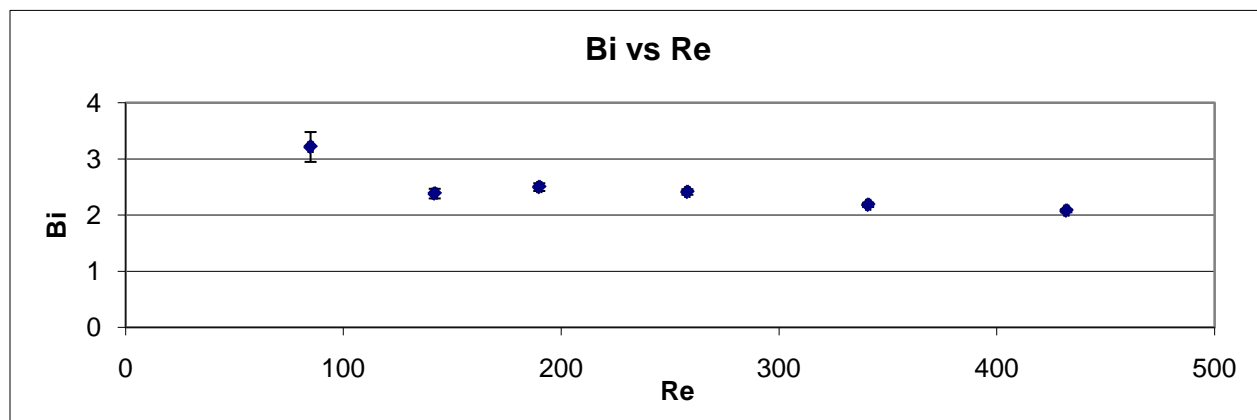
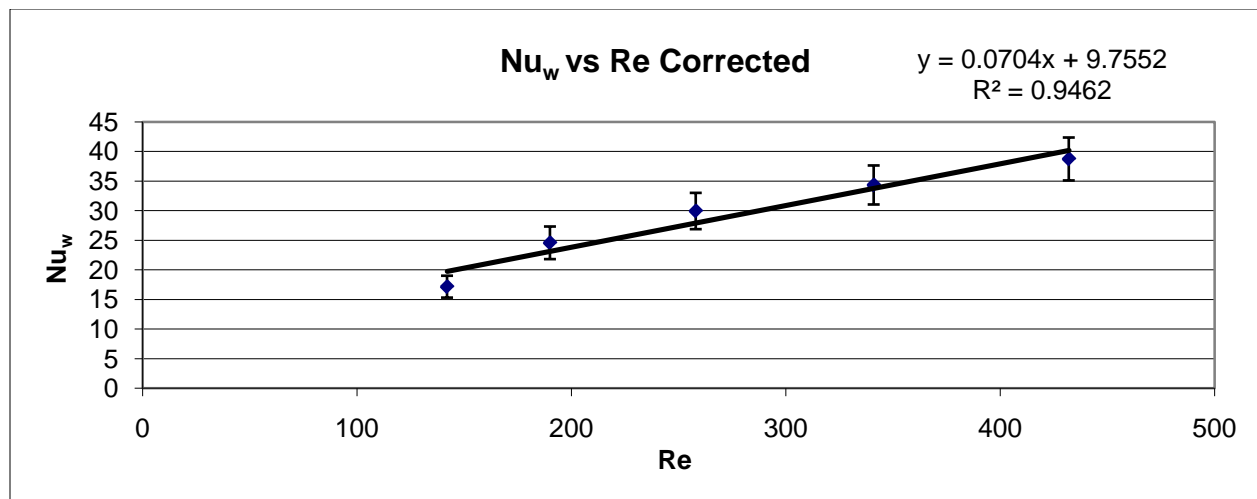


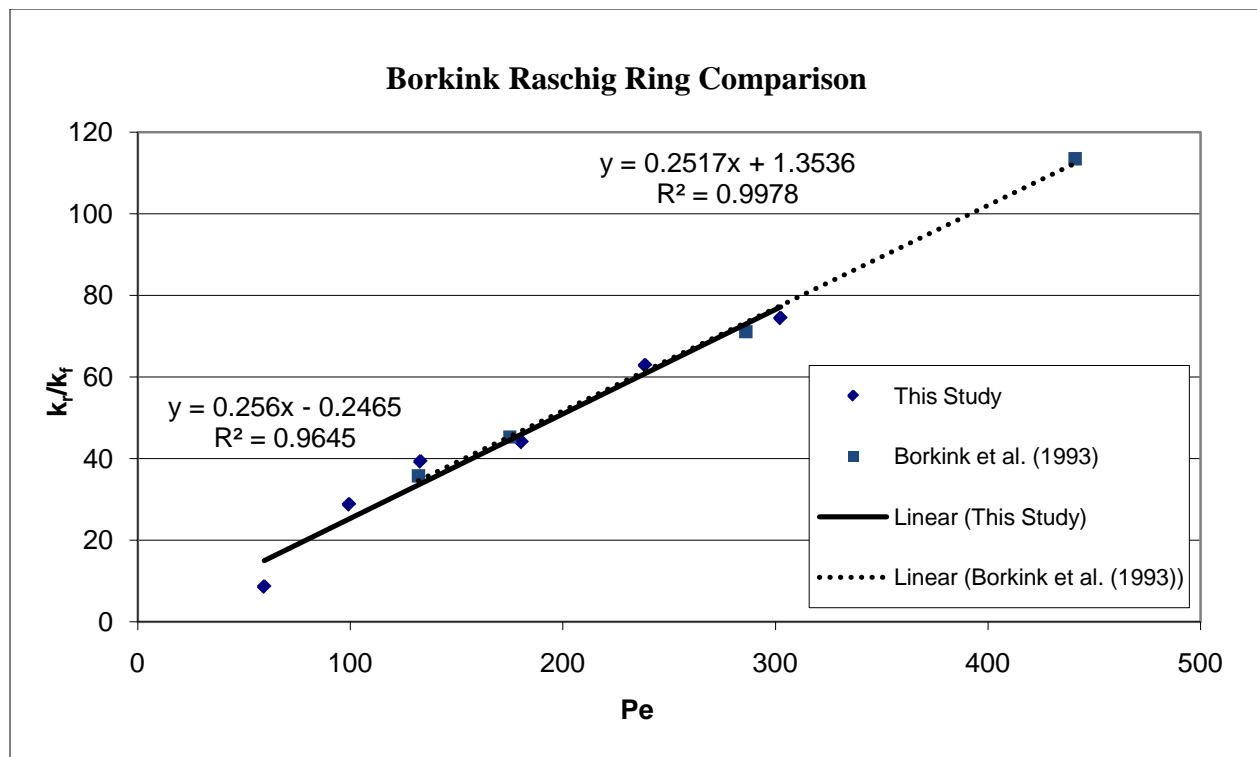
#### 4 inch column Spheres (Pollica)



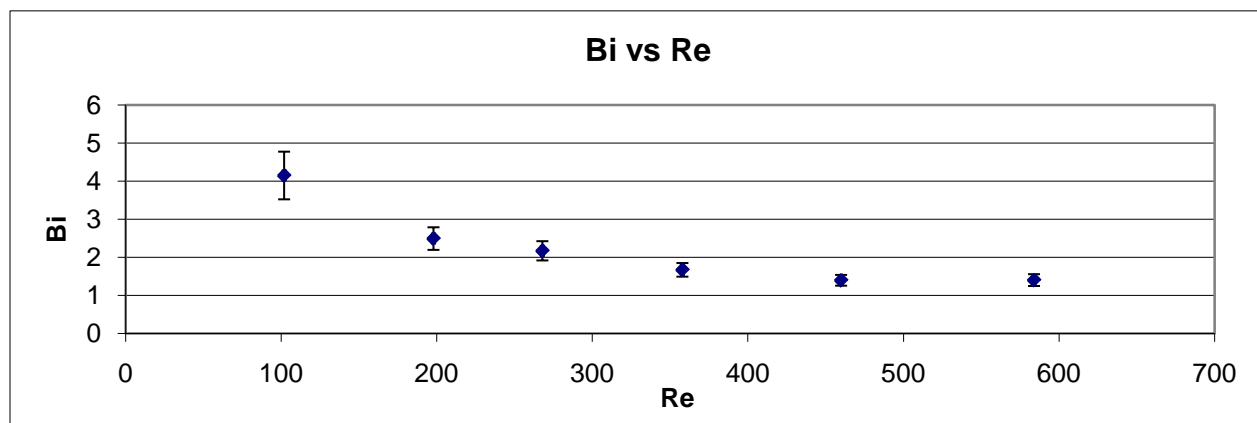
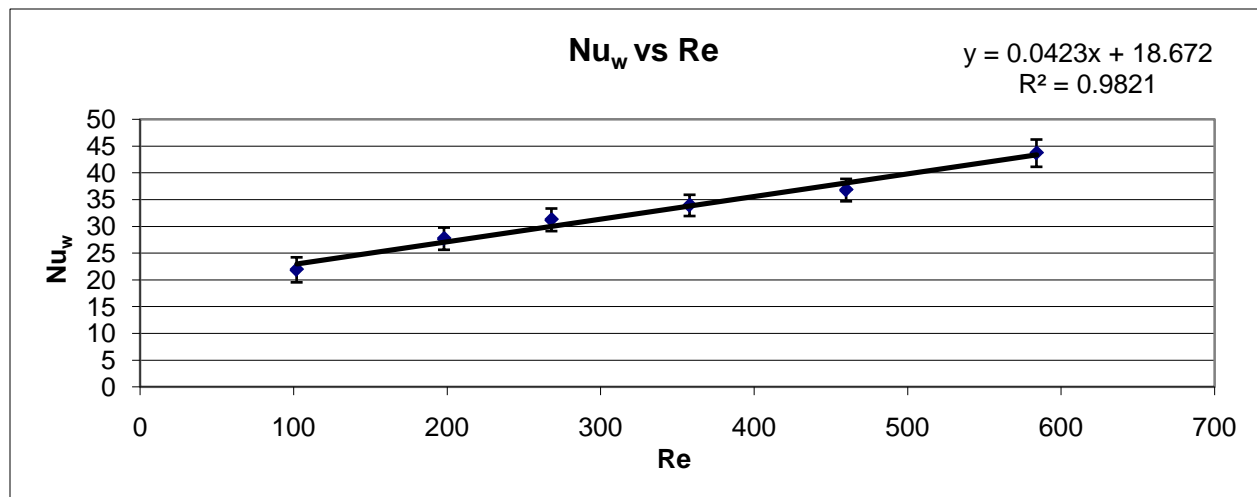
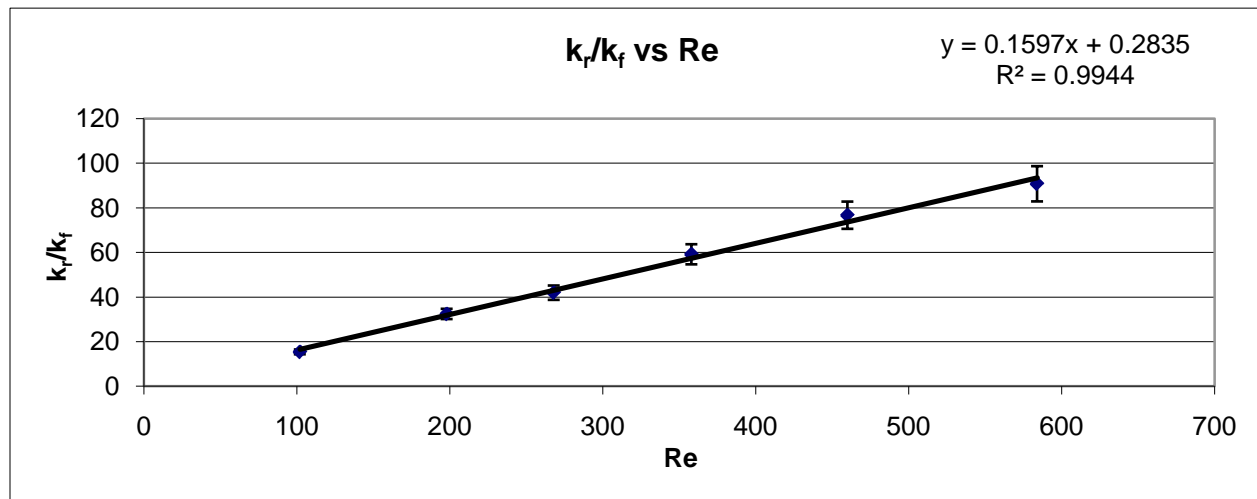
## 4 inch column Raschig Rings

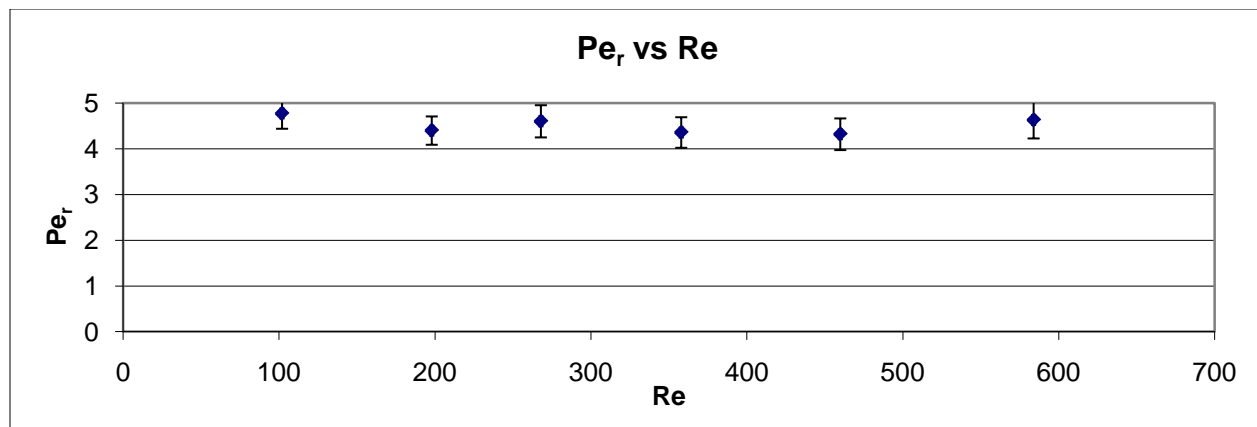




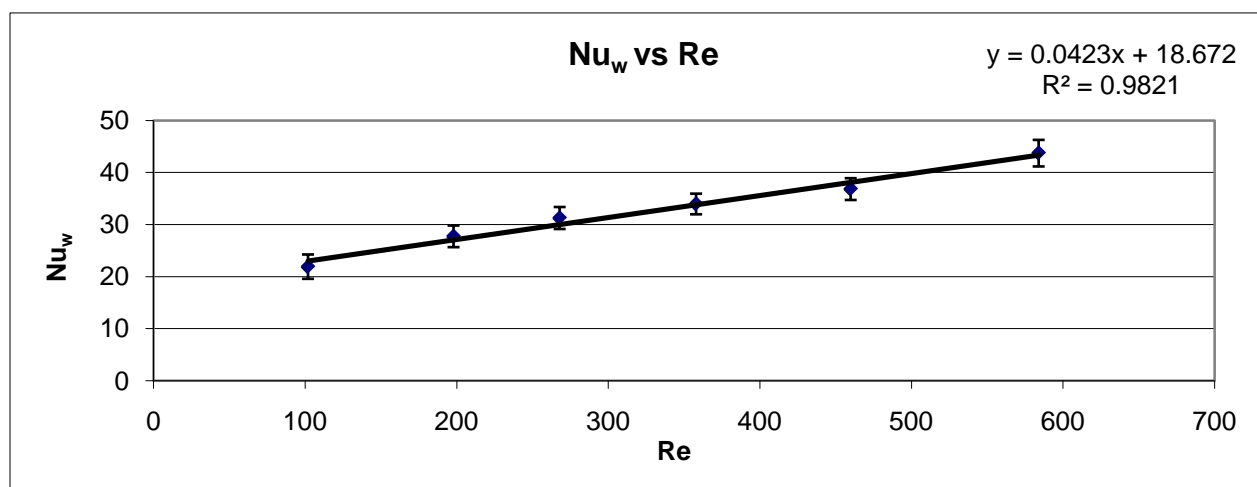
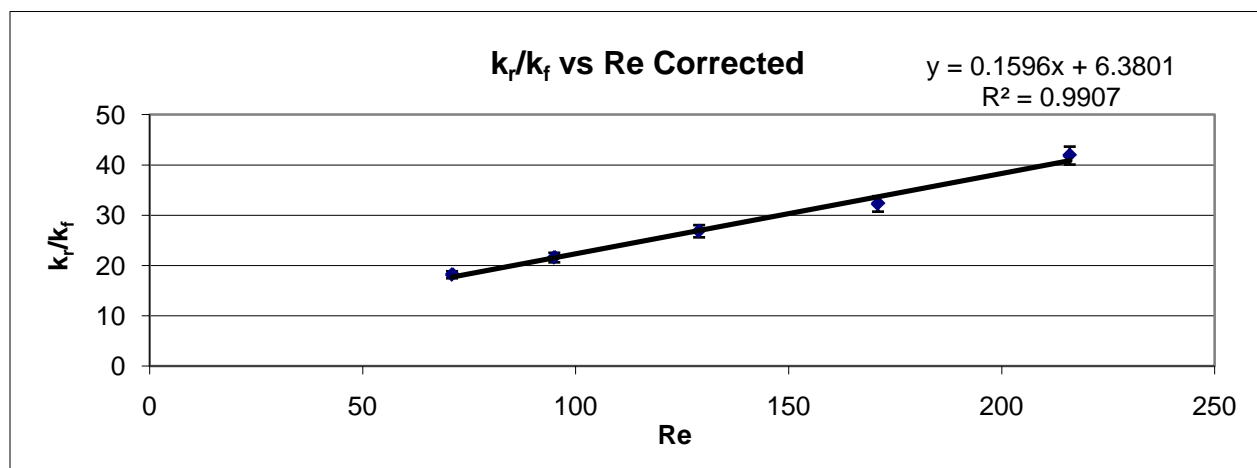
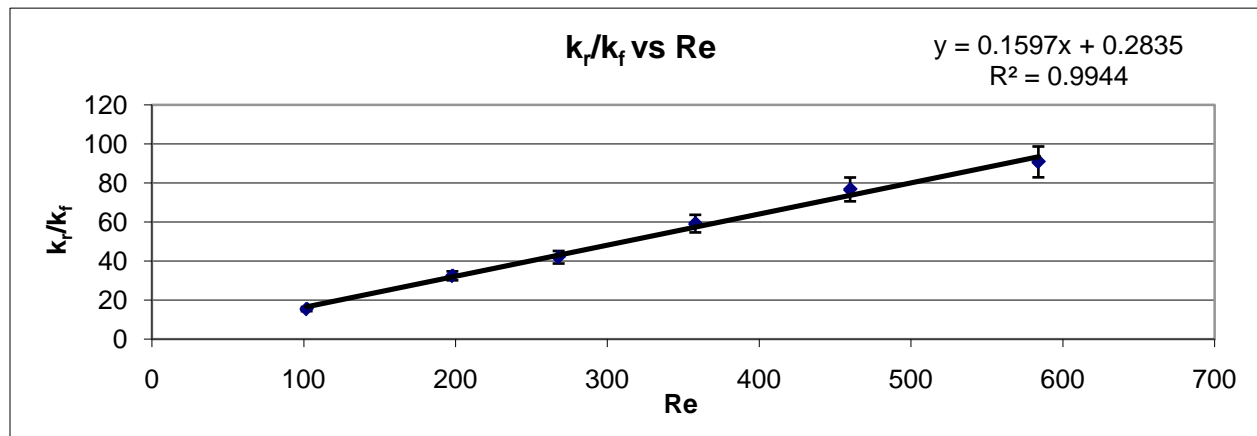


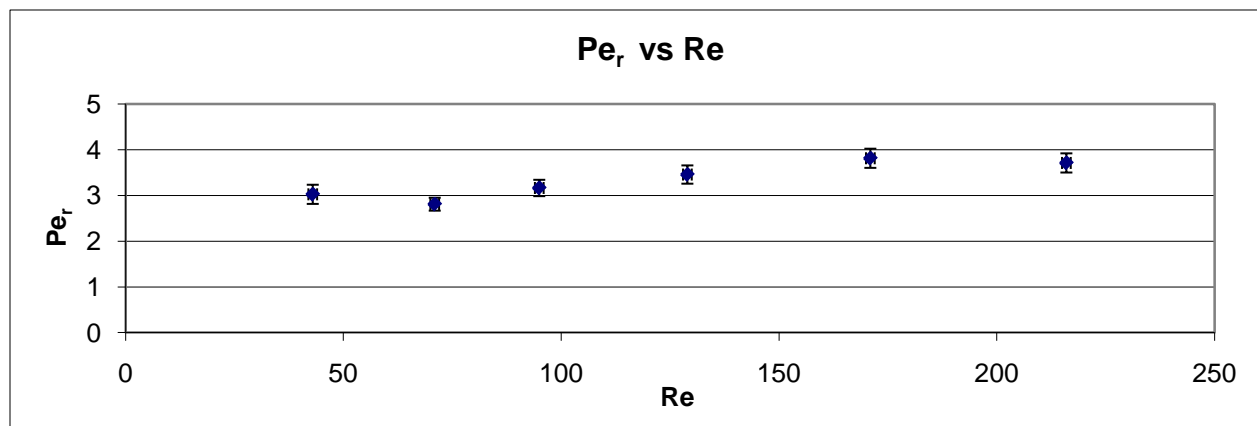
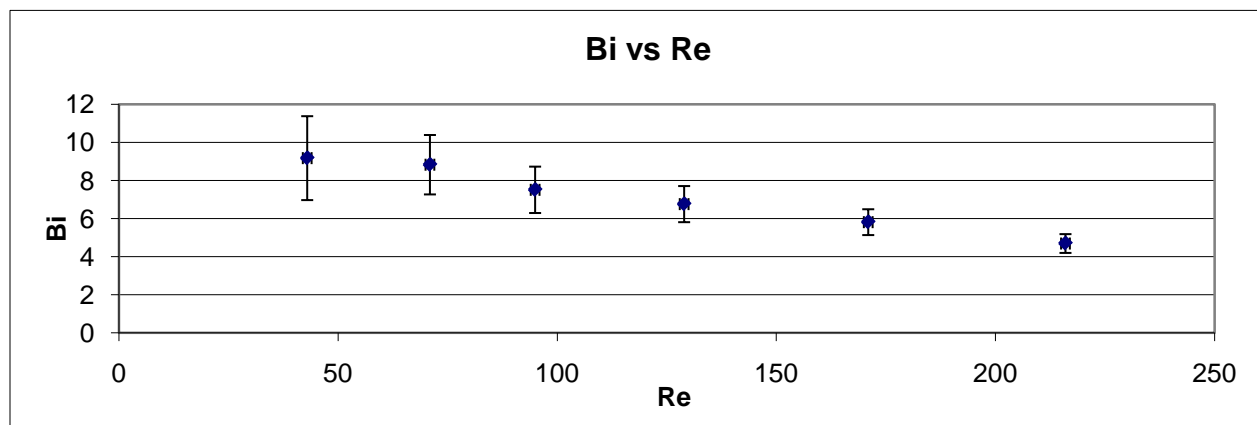
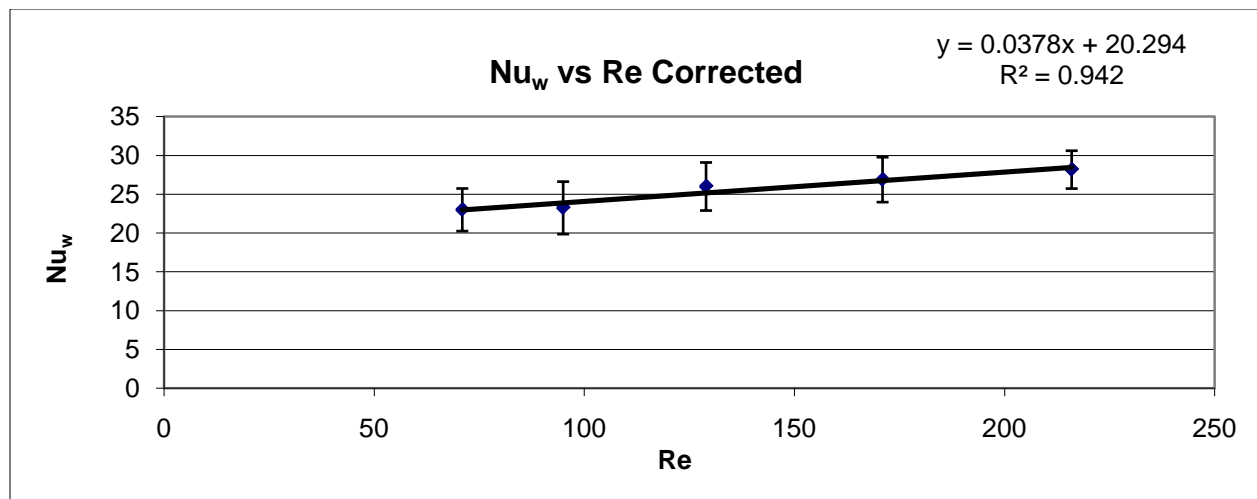
#### 4 inch column JM 4-hole cylinders





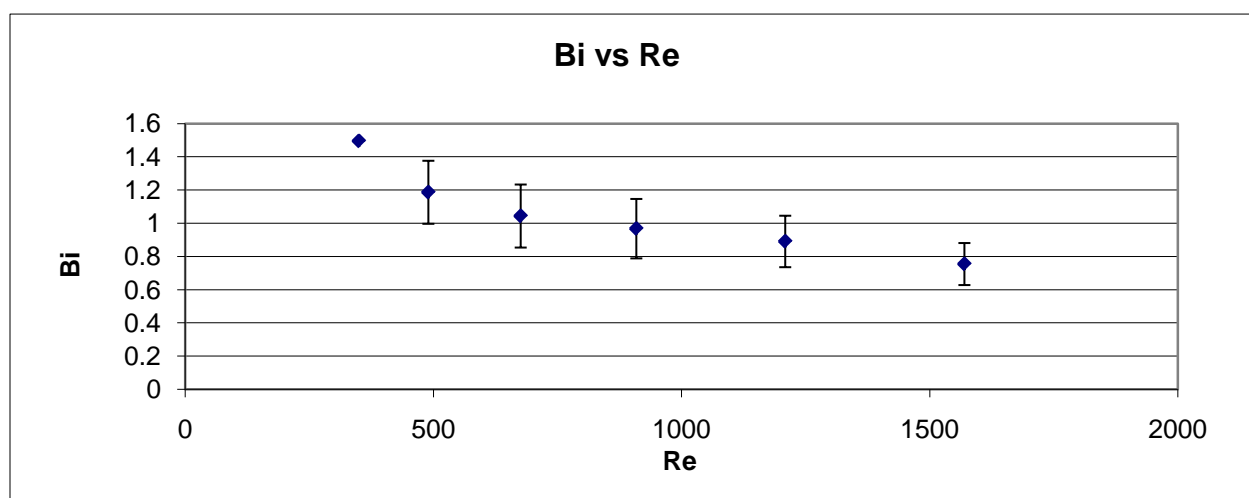
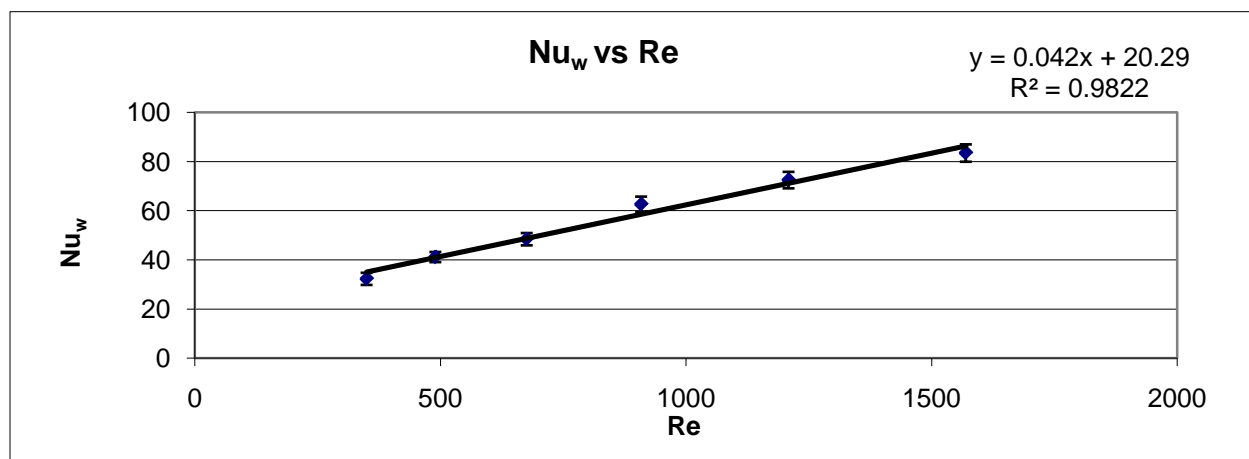
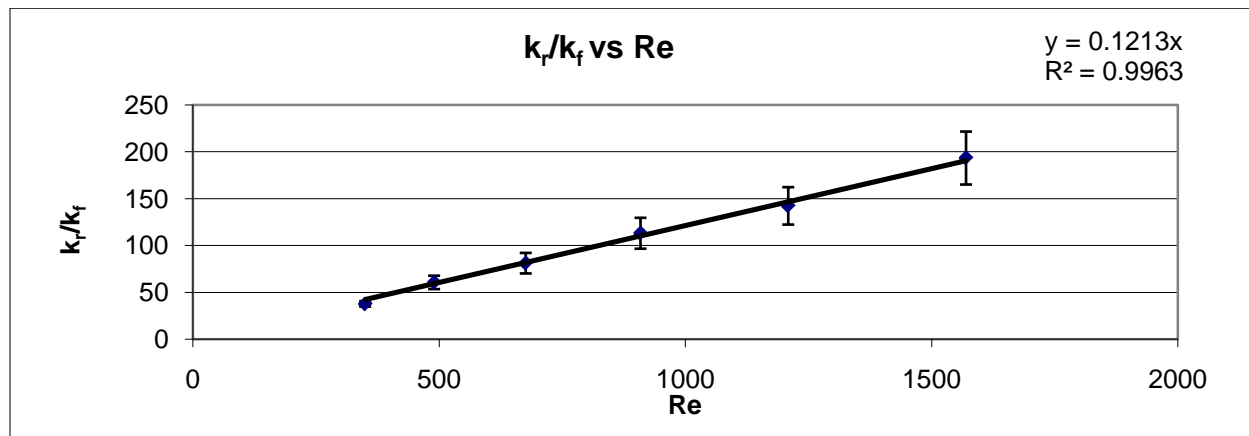
#### 4 inch column Monoliths

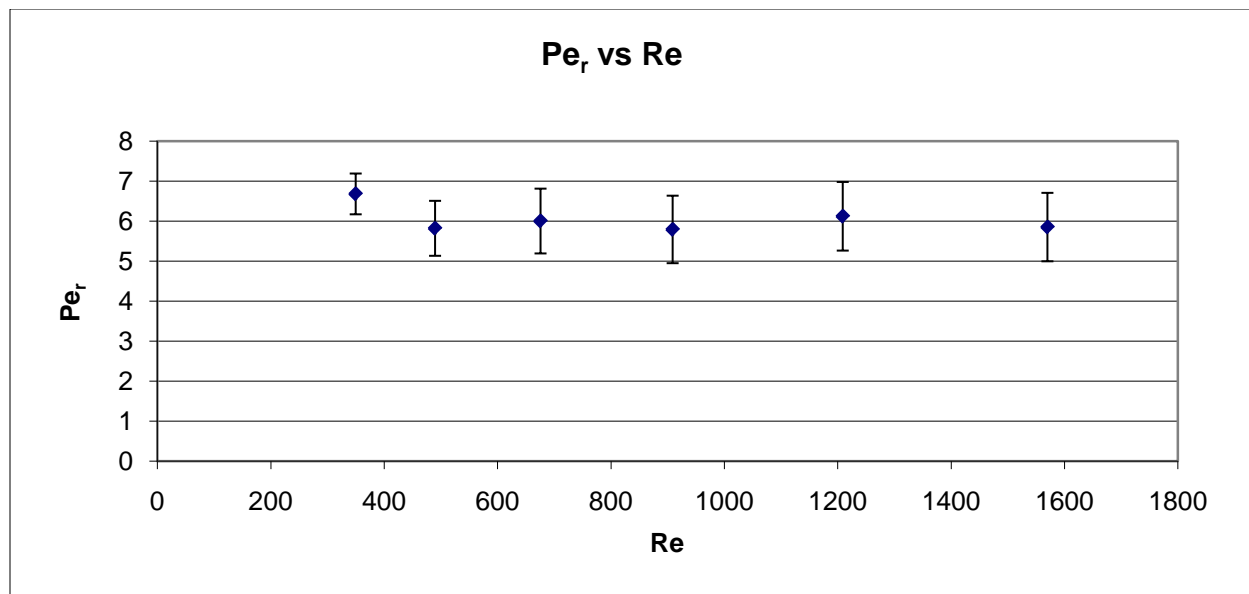




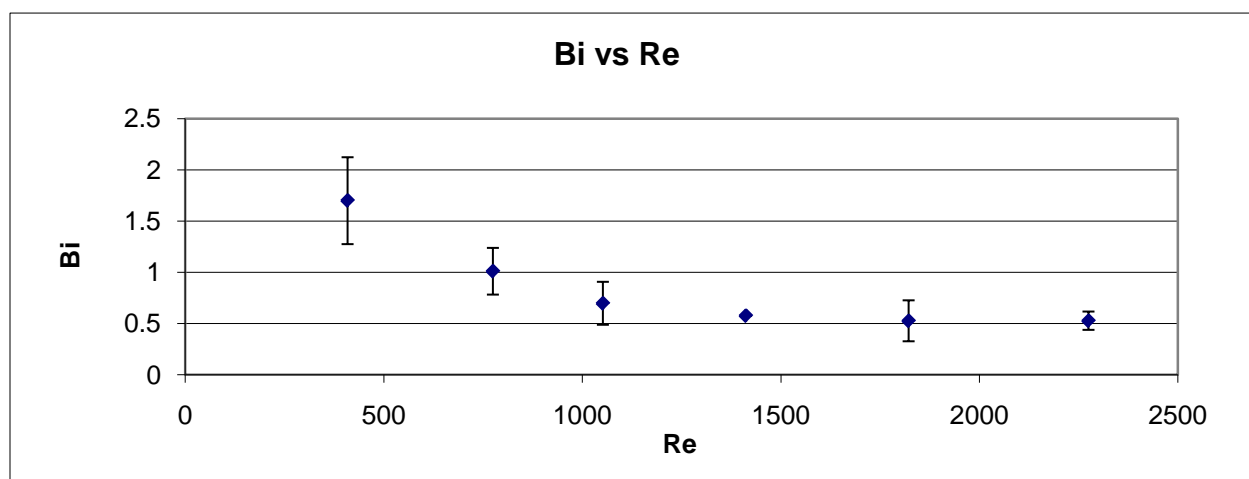
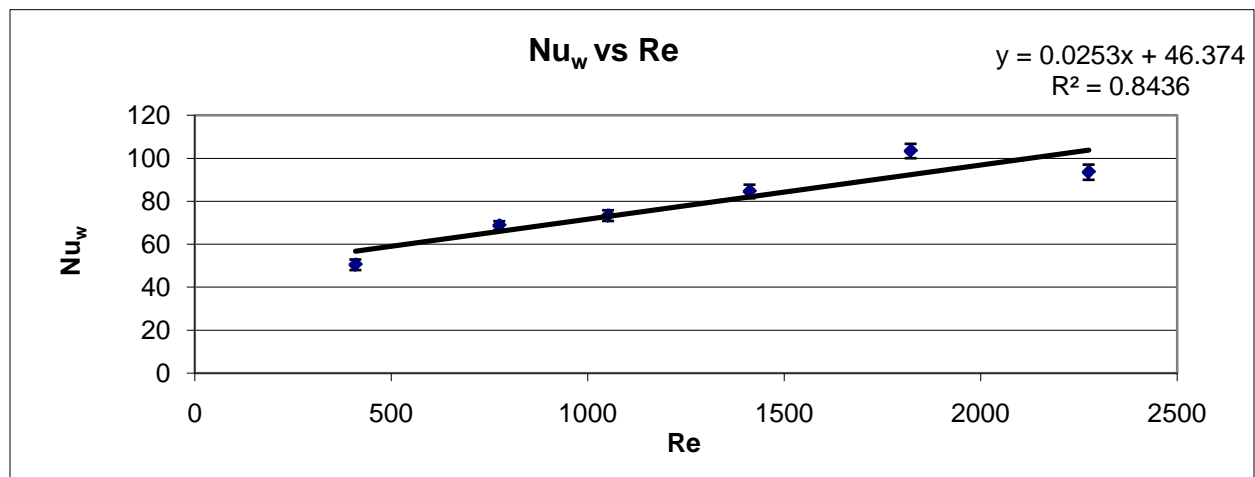
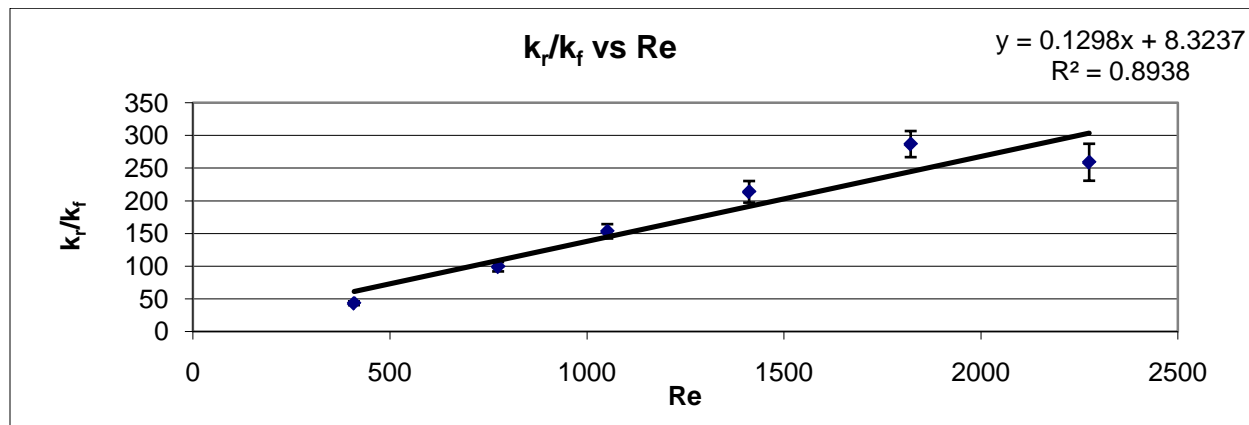


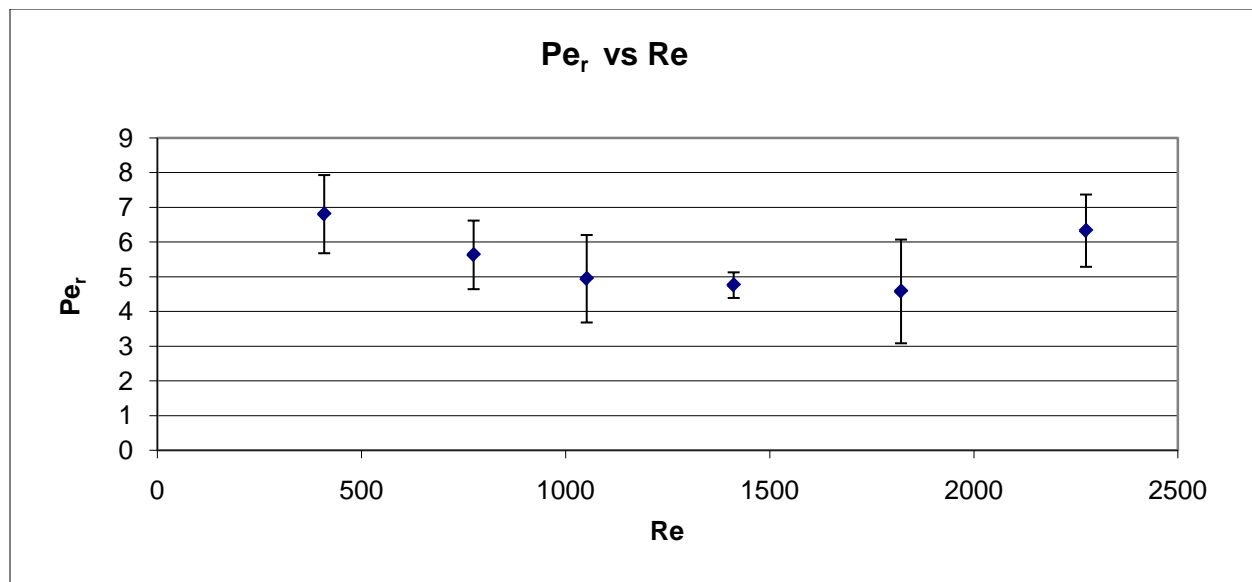
## 2 inch column Raschig Rings



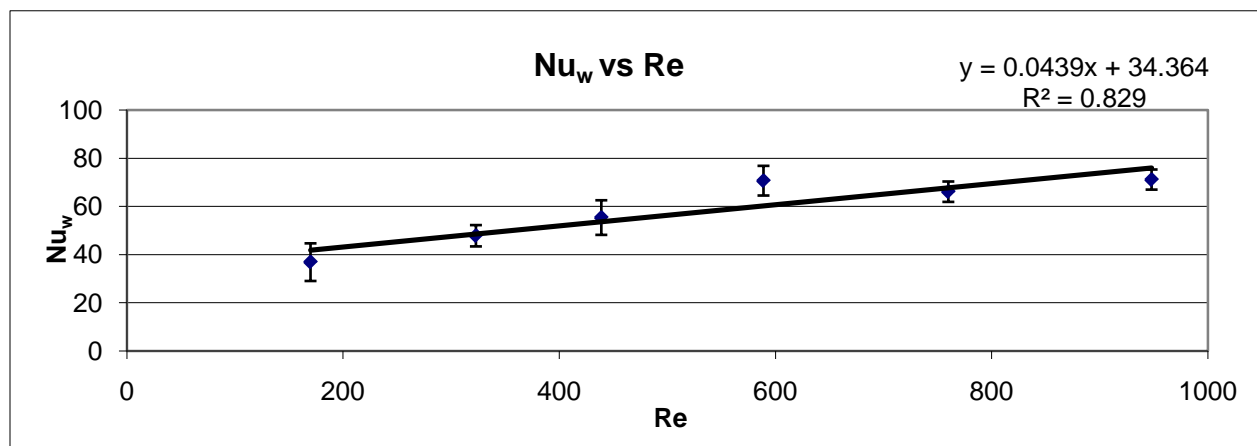
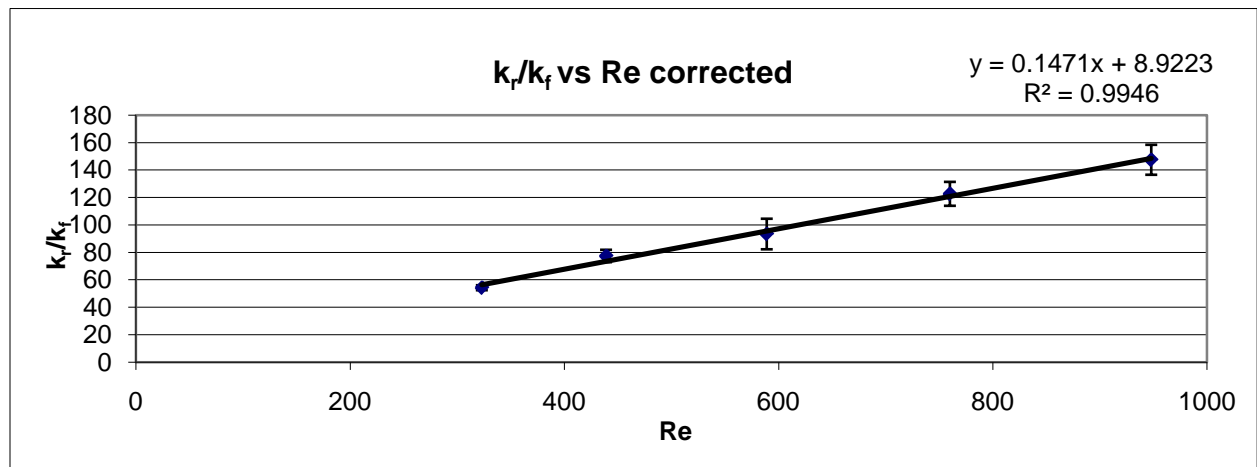
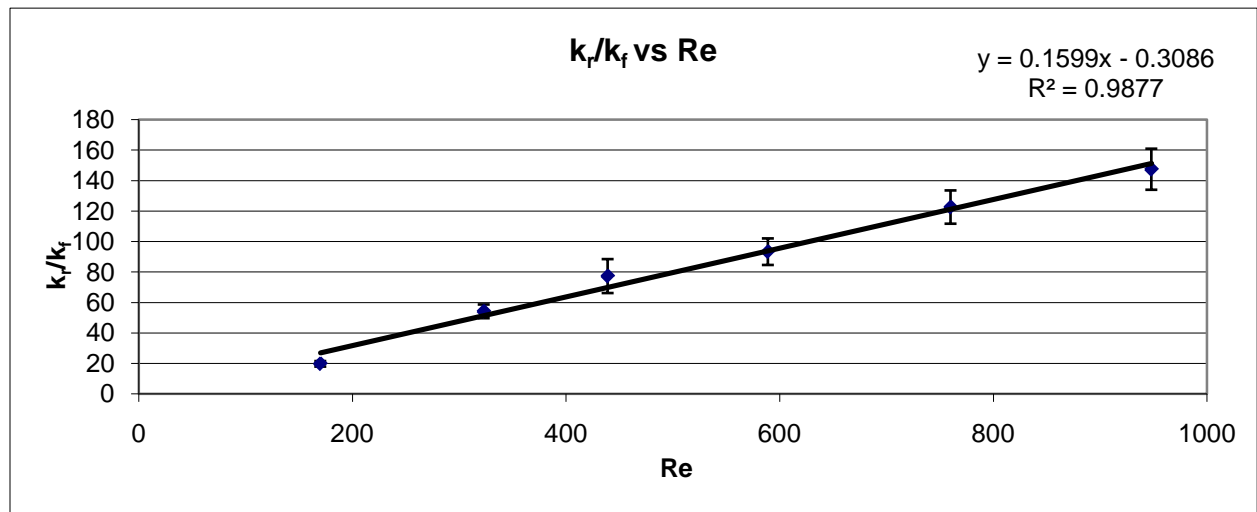


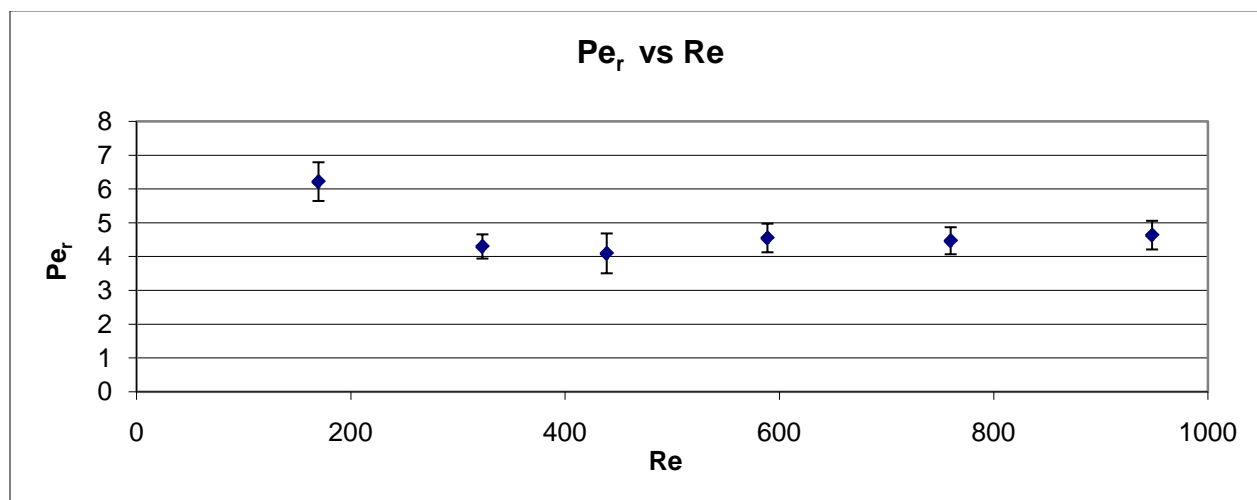
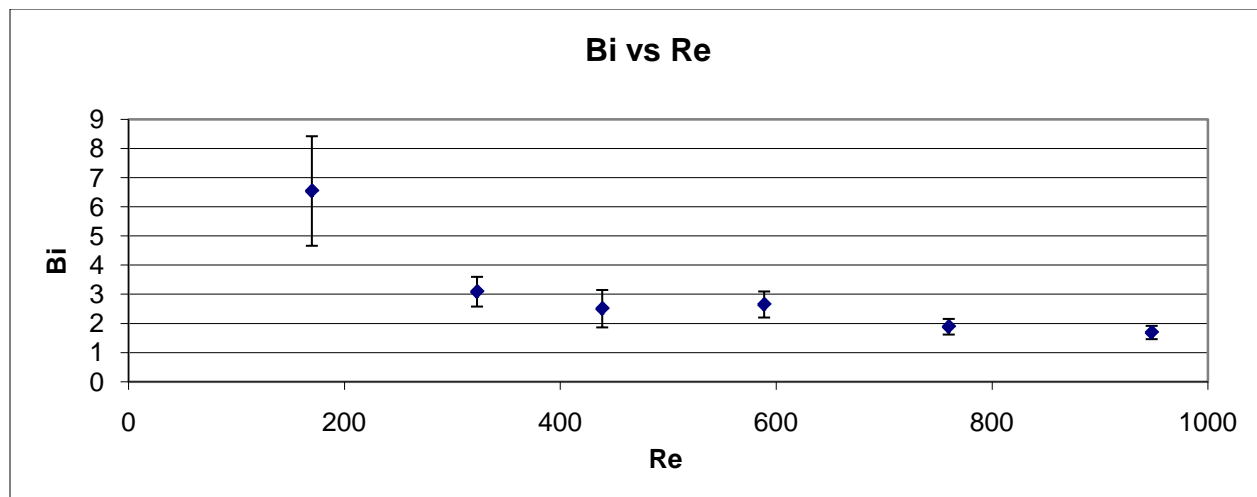
## 2 inch column 4-hole cylinders





## 2 inch column Monoliths





## Appendix F: F-test values

Column	Packing	Re	F/F95
4 Inch Column	Spheres	75	0.384578
		145	1.008367
		195	0.671051
		260	0.53057
		335	0.307174
		425	0.210865
	Raschig Rings	85	0.42928
		142	0.338531
		190	0.377891
		258	0.331282
		341	0.386067
		432	0.424467
	4-hole Cylinders	102	0.806354
		198	0.641854
		268	0.481158
		358	0.558235
		460	0.727119
		584	0.493383
	Monoliths	43	4.647282
		71	1.305055
		95	0.983003
		129	0.629038
		171	0.886185
		216	1.336643
2 Inch Column	Raschig Rings	350	3.912612
		490	2.507197
		676	3.358442
		909	5.43473
		1209	4.867726
		1570	3.886017
	4-hole Cylinders	409	0.82748
		775	2.000006
		1052	0.668281
		1412	1.207582
		1822	1.15873
		2275	1.615775
	Monoliths	170	2.37015
		323	3.674283
		439	10.36014
		589	2.966539
		760	0.986552
		948	1.847035